

COPPER AMALGAM -
SOME MECHANICAL AND
BIOLOGICAL PROPERTIES

by

GEORGE A. REID

Thesis presented for the degree of
Doctor of Philosophy
of the University of Edinburgh in
the Faculty of Medicine

APRIL 1973



ABSTRACT

Copper amalgam is still used today as a filling material for deciduous teeth. Good retentive properties backed by the bacteriocidal properties of the copper are claimed to reduce the incidence of secondary caries. The main disadvantages are green discolouration of the teeth restored by copper amalgam and unsustained reports of toxicity to the pulp.

The aim of this study was to investigate some of the mechanical properties of copper amalgam and carry out some biological experiments. Silver amalgam was used as a control material.

All the available data on the rather obscure metallurgy of copper amalgam has been presented. Only one intermediate phase, with a narrow band of homogeneity has been identified. It is an intermetallic or electron compound. Chemically such compounds are of closely controlled composition, usually involving small, simple ratios of the combining elements where laws of valency are not observed. Various formulae have been suggested. Physically such compounds are brittle and of variable hardness. It is thus distinguished chemically and structurally from the various types of dental silver amalgams.

Remarkably little data exists on physical properties; compressive strength (11 specimens), tensile strength (32), dimensional change (not known) and hardness (10). In this study over 100 specimens were

subjected to mechanical tests. It was found that copper amalgam exhibits approximately half to one third of the value for compressive strength and tensile strength of the values for dental silver amalgams used for comparison. Tests for these properties gave very variable results despite considerable efforts to standardise specimen production. These two properties do not appear to be related to residual mercury content. Dimensional change on setting was more consistent showing an overall average setting contraction of 0.13 per cent. The Vickers hardness of copper amalgams was similar to that of the silver amalgam the hardness value was significantly higher. The setting time of copper amalgam was undesirably slow.

The adaptability of copper amalgam was studied and compared with 5 different types of silver amalgam. (Only one previous worker has carried out such a study.) Copper amalgam adapted to the cavity wall better than the silver amalgams used for comparison. Scanning Electron Micrographs are included in this study and further demonstrate adaptation.

The effects of copper amalgam restorations and silver amalgam restorations on the dental pulps of rats were studied. Over 80 restorations of each amalgam were placed in the first maxillary molar teeth of rats, copper amalgam on one side of the maxilla and silver amalgam on the opposite side. There was no detectable difference between the effects of unlined copper amalgams and unlined silver amalgams on rat molar pulps over periods of 1 month

to 4 months even when the pulp had been surgically exposed. Copper amalgam restorations suffered more severe attrition than silver amalgams and were, therefore, less likely to be lost by accidental trauma. This finding explained the apparent good retentive properties of copper amalgam. Retention of the restoration was found to be a key factor in survival of the pulp.

Tissue culture studies demonstrated that in a controlled medium copper amalgam was a very much more effective source of copper and mercury ions (predominantly copper) than the pure metals themselves.

A model of the tooth/amalgam interface was designed to study the movement of bacteria in this area. By comparison with previous studies of this type the number of variables was reduced. It was found from 71 experiments using this model that a significantly higher number of experiments incorporating silver amalgam permitted bacteria to pass through the interface than the number with copper amalgam.

PART I	Page
1. Abstract.	i
2. THE AIM OF THE PRESENT STUDY.	3
3. PROPERTIES OF THE IDEAL FILLING MATERIAL.	4
4. REVIEW OF LITERATURE.	5
i. Historical.	
ii. Chemical and Metallurgical Survey.	9
iii. Physical Properties.	17
Summary	31
iv. Adaptation Studies.	32
v. Microleakage Studies.	38
Summary	50
vi. Effects on the Dental Pulp.	52
Summary	56
vii. Oligodynamic action.	57
Summary	63
viii. Microleakage studies using bacteria.	64
Summary	69
ix. Clinical Studies.	70
x. Mercury hazards.	71
Final Summary	73

CHAPTER 1

PART II	
5. PRELIMINARY STUDIES OF COPPER AMALGAM.	78
6. THE MECHANICAL PROPERTIES OF COPPER AMALGAM.	80
Method and Materials	80
Preparation of Specimens	80
Dimensional change - Apparatus	81
- Results	84
- Discussion	85

Compressive Strength - Apparatus	86
- Results	87
- Discussion	90
Vickers Hardness - Apparatus	91
- Results	92
- Discussion	93
Initial Setting Time - Apparatus	94
- Results	96
Summary of mechanical properties	96

CHAPTER 2

7. ADAPTABILITY AND MICROLEAKAGE STUDIES OF COPPER AMALGAM AND SILVER AMALGAMS	98
Adaptability - Method and Materials	98
- Discussion	101
Microleakage - Method and Materials	102
- Discussion	103
Conclusions	104
Photomicrographs	after 104

CHAPTER 3

8. A COMPARISON OF THE EFFECTS OF COPPER AMALGAM AND SILVER AMALGAM ON THE RAT MOLAR PULP	105
Method and Materials	106
Results	109
Histological Findings	114
Discussion	116
Conclusions	117
Photomicrographs	after 117

CHAPTER 4

9. TISSUE CULTURE STUDIES	118
Method and Materials	119
Results	121
Discussion	122
Conclusion	122

CHAPTER 5

10. BACTERIOLOGICAL STUDY	123
Method and Materials	125
Results	128
Discussion	128
Conclusion	131

CHAPTER 6

11. INVESTIGATION OF ERRORS	133
12. SUMMARY AND CONCLUSIONS	139
13. ACKNOWLEDGEMENTS	143
14. BIBLIOGRAPHY	144

"He (Mr. Turner) did not quite understand Mr. Fletcher's reference to Sullivan's stopping. He agreed with the chairman, that it was in many cases a most useful article. It could be used sometimes where it would be most hopeless to use any other stopping, and cases were continually coming under the notice of people in practice where this amalgam stopping had been used in a rude, rough manner by the old fashioned practitioners twenty, thirty or forty years ago. Why it should be so serviceable and still be of this porous character he was at a loss to tell. Certainly it had some chemical action upon the bone with which it came in contact, for if they cut out an old Sullivan's stopping, they would find the tooth in contact with it to be hard, while below might be soft; and this condition had been noticed elsewhere by Mr. Fletcher."

FLETCHER, T. 1895. Transactions of the Odontological Society, Vol. VII: 91.

INTRODUCTION

Copper amalgam, or Sullivan's cement, is used today in the conservation of deciduous molar teeth where conditions preclude ideal cavity preparation and the use of a "dry-field" technique. Copper and its derivatives have always been associated with germicidal properties and copper amalgam has a reputation among children's dentists for retaining well and preventing the formation of secondary caries.

Discolouration of the restoration and green staining of the restored teeth would appear to be the main disadvantages and are perhaps of little significance in the deciduous dentition. Over long periods copper amalgam restorations exhibit what is known as "cupping". This is due to corrosion, causing loss of weight in the restoration and ultimate failure.

The popular theory advanced, but as yet unsustained, for the apparent prevention of secondary caries is that the oral fluid penetrates the interface between the cavity wall and the restoration as a result of the setting contraction of copper amalgam and, combined with the products of corrosion, inhibits caries formation by destroying either the cariogenic bacteria or their nutrients.

THE AIM OF THE PRESENT STUDY

To investigate some of the mechanical and biological properties of copper amalgam.

To provide data for its assessment as a dental restorative material.

To compare copper amalgam with silver amalgam as the reference filling material.

A dental restorative material is a material which replaces lost tooth substance in form and function. In the past any irritant or toxic effects of dental restorative materials on the dentine-pulp unit have been accepted since separate pulp protection can be provided by a non-irritant cavity lining. In deep cavities under metal restorations, such a lining would also serve as thermal insulation.

THE IDEAL FILLING MATERIAL

The ideal dental restorative material should have the following properties:-

- (1) It should not be affected by the fluids in the mouth.
- (2) It should adapt closely to the cavity wall to form a marginal seal.
- (3) It should form an adhesive bond with the cavity wall.
- (4) It should not harm the dental pulp or oral tissues.
- (5) It should have a coefficient of expansion and thermal conductivity similar to that of tooth substance.
- (6) Its crushing strength should be sufficient to withstand the stresses of mastication.
- (7) Its hardness and abrasive resistance should be sufficient to prevent damage during function.
- (8) The technique of preparation and insertion of the material should be simple.
- (9) When the restoration is extended it should be capable of providing a chemical bond in the mouth at original strength.
- (10) It should match tooth substance exactly in colour and texture.

In 1964 and 1965, Jonathan Baskin published a book in Latin under the title of "The History of the World" (1964) and "The History of the World" (1965). The book is a history of the world from the beginning of time to the present. It is a very good book and is highly recommended.

The book is a history of the world from the beginning of time to the present. It is a very good book and is highly recommended. The book is a history of the world from the beginning of time to the present. It is a very good book and is highly recommended.

Therefore, the book is a history of the world from the beginning of time to the present. It is a very good book and is highly recommended.

The book is a history of the world from the beginning of time to the present. It is a very good book and is highly recommended.

THE REVIEW OF LITERATURE

The book is a history of the world from the beginning of time to the present. It is a very good book and is highly recommended. The book is a history of the world from the beginning of time to the present. It is a very good book and is highly recommended.

The book is a history of the world from the beginning of time to the present. It is a very good book and is highly recommended. The book is a history of the world from the beginning of time to the present. It is a very good book and is highly recommended.

HISTORICAL REVIEW

In 1634 and 1657 Joannes Stocker published a book in Leiden under the title of "Praxis Aurea ad corporis humani morbos." This book contained the following formula:

"Dissolve vitriol with a strong acid in a bowl and add mercurium. Boil this substance and the mercurium will change into an amalgam. Put this in the cavities of the teeth; it gets hard as stone and keeps in every cavity".

De MAAR (1968) states that copper amalgam must, therefore, have been known in Holland at the beginning of the 17th century.

Copper amalgam also appears to have been a modelling material known as Viennese Metal Cement or Metallic Mastic, used in the reproduction of ornaments.

HENLEY (1965) According to De BRAY (1856):

"When a medal, obtained with an amalgam of 45 per cent of copper by compression in the soft state in moulds of gutta percha, is heated progressively to redness in an atmosphere of hydrogen, the quicksilver is volatilized gradually and the particles of copper come together without fusion in such a way as to produce a faithful reproduction, formed exclusively of metallic copper, of the original medal. When used as a cement if applied while

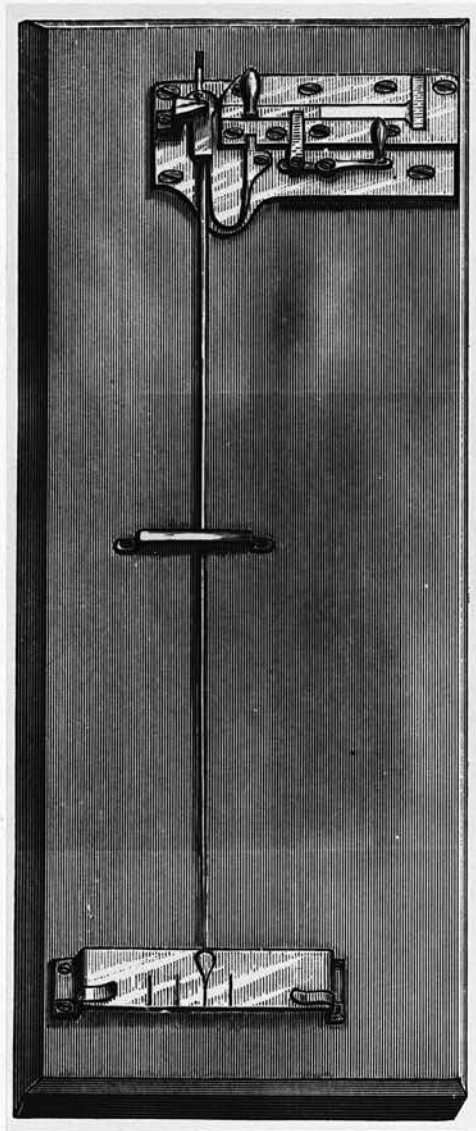


Fig. I

Apparatus for measuring the dimensional change on setting of dental amalgams from 'Plastics and Plastic Filling' by J. Foster Flagg (1891) Philadelphia.

6

hot and plastic to the deoxidized surface of two pieces of metal, these latter will unite so firmly that in about 10 to 12 hours the metal may be subjected to any mechanical process. The properties of this composition render it very useful for various purposes, and it forms a most effective cement for fine metal articles which cannot be soldered in fire."

TOMES, J. (1861) packed what he termed "pure copper amalgam" and Sullivan's cement into small moulds and "observed" the margins. He did not define the difference between the two copper amalgams nor did he describe the apparatus which he used for measurement. However FLAGG (1891), in his book "Plastics and Plastic Filling", illustrates the apparatus which might possibly have been used by Tomes, Fig. (1). The fact that Tomes found that his "pure copper amalgam", unlike Sullivan's cement, did not show any contraction, casts some doubt on the accuracy of his experiment.

FLETCHER (1875) sought to demonstrate marginal leakage by packing amalgams into glass tubes and immersing the tubes in a dye. He assumed that the presence of dye between the amalgam and the walls of the glass tube signified a decrease in the dimensions of the amalgam and on this basis he found

that all amalgams tested showed a setting contraction after insertion. He made no allowance for the considerable dimensional variation between amalgam and glass due to thermal changes during the course of the experiment, thus his conclusions are suspect.

TOMES, C. (1875) without describing the experimental method, measured the specific gravity before and after setting and found that all amalgams contracted.

In Dental Cosmos (January 1891) one section of operators condemned copper amalgam thus:

"No antiseptic properties, soluble in certain mouths, detached particles, discoloured other teeth, danger of absorbtion, subject to shrinkage allowing moisture to penetrate the tubules, and the tendency to soften the cervical.

.

Leakage was present only where packed too soft or too wet. Discolouration resulted from three causes:

1. Defective manipulation.
2. Teeth of poor structure which were permeable to water, resulting in oxidation which is beneficial to the tooth.
3. Impure preparation."

These attempts to evaluate the performance of amalgam on the bench were based on clinical experience and information of anecdotal quality. Unfortunately insufficient evidence was presented to sustain the observations but clearly the aim was to provide some standard by which the numerous filling materials of the time could be judged.

CHEMICAL AND METALLURGICAL SURVEY

Dental copper amalgam is supplied by the manufacturers as a ready-made amalgam. It is heated for use until beads of mercury form on the surface of the pellet. It is rendered completely plastic by trituration in a mortar and pestle and returns to room temperature at this stage. Excess mercury is squeezed off and thereafter manipulation is the same as for silver amalgam.

Dental silver amalgam, made from mechanically divided silver/tin alloy combined with mercury immediately prior to use to become an aggregate or particle/matrix system differs from dental copper amalgam which, for dental use, is made chemically from copper precipitate and mercury although they perform the same function as restorative materials and are manipulated in the mouth by the same technique.

The heating prior to trituration is a characteristic peculiar to copper amalgam. COGGAN (1933), WORMINGTON (1939) and SCHARFENBERG (1948) found that the temperature to which the copper amalgam was heated affected the physical properties.

COMPOSITION

SKINNER (1946) gives the following table for the compositions of dental copper amalgams

	Mercury %	Copper %	Other Metals %
A.	66.3	32.5	Zn, 0.7
B.	61.7	37.1	Zn (not determined)
C.	67.8	32.2	Trace O ₂
D.	72.1	26.6	Zn (not determined)
E.	70.5	28.2	Zn (not determined)
F.	62.1	37.6	
G.	64.1	35.7	
H.	67.7	30.7	Sn, 1.5% approx.
I.	69.3	28.5	Sn, 2% approx.

after SKINNER (1946)*

GRANATH (1961) gives mean values of 65.3 and 63.8 per cent for the weight of mercury in 10 specimens of two brands of copper amalgam prepared for hardness tests. No data are given for their composition prior to preparation.

No data on the composition of completed copper amalgam fillings is available in the literature.

*Data provided by H. K. WORNER, Professor of Metallurgy, Melbourne University.

THE SOLUBILITY OF COPPER IN MERCURY

The degree of solubility of the metal in mercury has been used as one of the criteria in the classification of amalgams (HOHN, 1950). He divides amalgams into three different groups according to their technical applicability. Metals having good solubility are close to mercury, horizontally and vertically, in the periodic classification of the elements and indicates that there is a correlation between solubility and the structure of the outer electron shells.

The solubility of copper in mercury at room temperature is low. According to HANSEN (1958) it is 0.003 per cent by weight. KATOH (1929) assumed that mercury is not soluble in copper, since the lattice parameter for the copper phase in copper amalgam is the same as that of pure copper. The solubility of silver in mercury is 0.035 per cent by weight at 20°C; the corresponding value for tin is 0.6% by weight at 15 - 18°C (HANSEN, 1958).

THE PHASE DIAGRAM

Little is known about the solid state of copper amalgam. TAMMAN & STASSFURTH (1925) carried out a microscopic and thermal study by noting temperatures at which beads of mercury appeared on heating copper amalgams of varying composition. They suggested the hypothetical phase diagram shown in Fig. 2. According to this diagram there should be

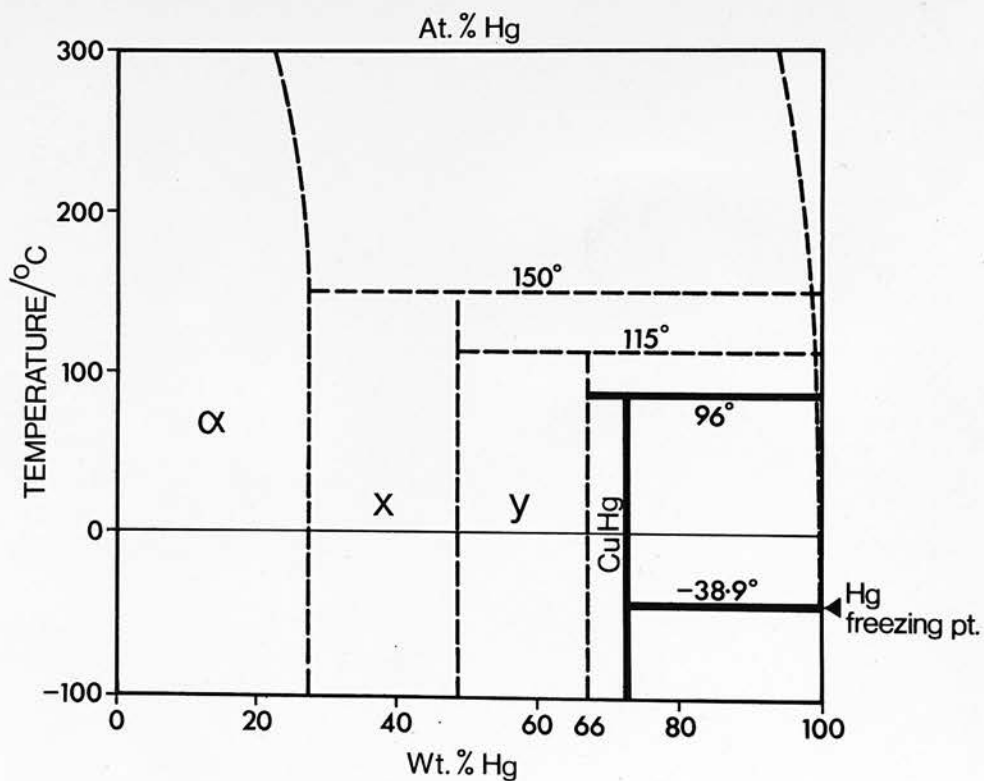


Fig. 2

Phase diagram for copper amalgam after Tamman and Stassfurth (1925).

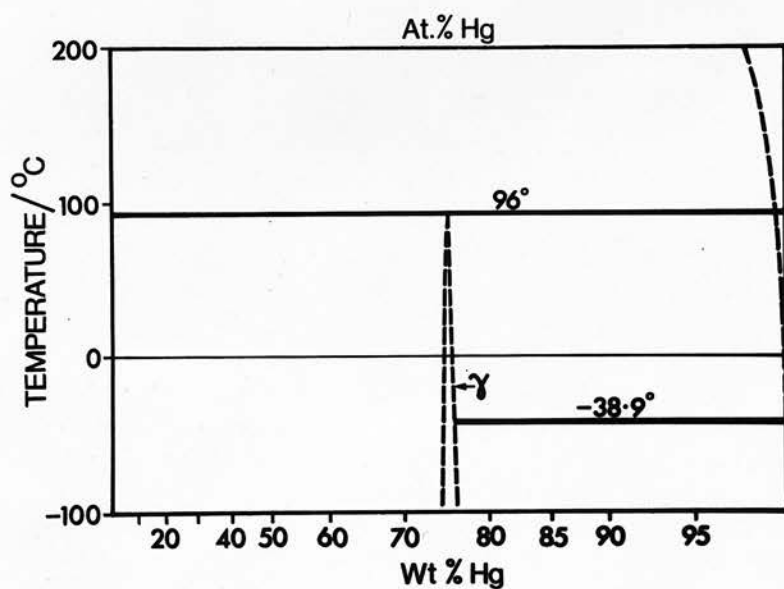


Fig. 3

Phase diagram for copper amalgam after Hansen (1958).

three intermediate phases which decompose at 150, 115 and 96°C but only one of these (96°C) has been shown to exist with certainty (CHAO and COSTA, 1968).

X-RAY DIFFRACTION STUDIES

X-ray diffraction studies have demonstrated the presence of a single phase with a structure similar to that of gamma-brass (D82 type) and a very much narrower field of homogeneity than the nine atomic per cent solid solution of mercury in copper suggested by Tamman and Stassfurth (HANSEN, 1958, Fig. 3).

ELECTRON COMPOUNDS

SMALLMAN (1970) distinguishes between two types of intermediate phase; those which exist over wide ranges of composition known as secondary solid solutions and those having a small range of homogeneity known as intermetallic compounds.

BOWDEN (1950) mentions some characteristics of intermetallic compounds:-

"(a) A metal never combines with a metal in its own subgroup of the Periodic Table. Copper, for example, does not form intermetallic compounds with silver or gold.

(b) The ordinary valency rules are not obeyed and formulae such as Pb Mg , Cu Mg_2 , Ag Mg_3 and Au_2Mg_5 are quite common.

- (c) Intermetallic compounds are usually hard and brittle.
- (d) Intermetallic compounds are poor conductors of heat and electricity.
- (e) X-ray analysis reveals that the various atoms are arranged in a characteristic, ordered space lattice.
- (f) The chemical reactivity of an intermetallic compound may be greater or less than that of the constituent metals. The compound Mg_2Pb oxidises spontaneously at ordinary temperature, but there are other intermetallic compounds which are more inert than the parent metals."

The factor governing the composition of intermetallic compounds of copper, silver and gold was perceived by HUME-ROTHERY (1926) to be the ratio of the number of valency electrons to the total number of atoms of both kinds in the unit of formula. Hence intermetallic phases of this kind are called electron compounds. For gamma-brass structures this ratio is 21: 13 and would lead to the formula $\text{Cu}_5 \text{Hg}_8$ for copper amalgam (13 atoms and $5 + 16 = 21$ valence electrons) but an important dimensional factor displaces the stability zone towards a lower proportion of mercury (HUME-ROTHERY et al (1951). According to HANSEN (1958) the most likely formulae are $\text{Cu}_4 \text{Hg}_3$ and Cu Hg .

LINDAHL et al (1968) suggest that the calculated density for $\text{Cu}_{15} \text{Hg}_{11}$, which approximates closely to one of the two possible analytical formulae, $\text{Cu}_4 \text{Hg}_3$, corresponds most accurately to their own result of 12.6 obtained by weighing electrolytically prepared copper amalgam in air and in chloroform. They state that the alternative analytical formula, Cu Hg , would produce a calculated density of 13.7.

CHAO and COSTA (1968), in electrolytic studies, concluded that "the solid phase of freshly prepared copper amalgam consists of copper crystals which slowly combine with mercury to form a bimetallic compound. The total speed of formation of this compound corresponds to the crystallisation or "ageing" of the compound, which explains the existence of a zone of over-saturation of copper in the amalgam with the slow appearance of solid crystals of copper amalgam."

PREPARATION

Copper amalgams may be prepared by many methods, but the most common involves electrolysis of aqueous solutions of copper salts using mercury as a cathode (LIHL, 1953).

Copper amalgams have also been prepared by displacement of lead or bismuth in mercury, using copper prepared by displacement from acidified copper sulphate solution by zinc, (IRVINE and RUSSELL, 1932).

DOW (1963) prepared copper amalgam by coating copper with lead or tin, then agitated the coated copper with mercury in an acid solution of the intermediate metal (i.e. lead or tin).

The simplest laboratory method is that of TERREY and WRIGHT (1928). Copper sponge, prepared by displacement from acidified copper sulphate solution by zinc, was mixed with mercury under dilute acid conditions. The amalgam was pressed free of acid, washed with distilled water, and left to harden at room temperature.

MELTING POINT

A melting point of 135°C was quoted for copper amalgam by GUNZ and DE GREIFT (1912). SCHARFENBERG (1948) found that the lowest temperature to which copper amalgam could be heated for trituration was 150°C . HUTCHESON (1969) used a Dupont Differential Thermal Analyser to study the behaviour of copper amalgams as they underwent physical and chemical changes during heating and cooling. The analyser consists of a thermocouple attached to the specimen to be examined and a similar thermocouple attached to a reference material which does not exhibit a thermal transition over the same temperature range. Any difference in temperature between the two thermocouples can be measured on a voltmeter. A third thermocouple was used to measure the environmental

temperature and control the rate of heating and cooling in the experiment. Thermograms were obtained by plotting time against temperature. His results were as follows:

Copper amalgam	Temperature of Transition on heating
Ash's Globe (new formula)	157°C
Cupro Muc (Merz and Co.)	163°C
Lab. preparation 40% Hg	137°C
Lab. preparation 75% Hg	153°C
Lab. preparation 80% Hg	155°C

after HUTCHESON (1969)

All the thermograms exhibited an endothermic peak on heating only. Hutcheson concluded that the absence of cooling peaks indicated irreversible reactions, the nature of which could not be identified. These were the only thermograms attempted. Variational errors were not discussed.

PHYSICAL PROPERTIES

It was not until 1928 when the American Dental Association, working under a co-operative agreement with the National Bureau of Standards, made any attempt to produce a standard for dental amalgam. TAYLOR (1929) in his report to this Commission entitled "A Survey of Amalgam Alloys" makes the following observations on copper amalgam:

"Copper amalgam shows a continuous shrinkage for a period of twenty-four hours, although the total change is less than in many of the unsatisfactory high silver alloys, their flow values are satisfactory, their crushing strength values are high but erratic, owing perhaps to a difficulty in heating the material uniformly in amalgamating them according to the directions furnished. The edges chip easily and the alloys do not machine well. The manipulatory methods used in handling copper amalgams are such that it is very difficult to produce two samples from the same alloy having the same or nearly the same physical properties. We may therefore expect a wide variation in strength, setting time and other characteristics in our finished products."

He gave no details of the composition or number of the copper amalgams tested but in a table of "Physical Properties of Amalgam Alloys", he gave the data as follows:

Manufacturer and Brand	Contraction % to 1st minimum	Total change % in 24 hr.	Flow % 250 kg. /cm ²	Crushing strength lb. per sq. inch (N/mm ²)
S. S. White Copper Amalgam	-0.06	-0.06	0.1	48,000 (331)
L. D. Caulk Copper Amalgam	-0.015	-0.015	0.1	44,000 ¹ (303)
W. V-B Ames Co. Copper Amalgam	-0.048	-0.048	0.1	38,200 (263)

adapted from TAYLOR (1929)

¹Other values as low as 18,100 lb. per square inch 124 N/mm² depending on the mercury content (Taylor's note).

Taylor's results were obtained by the use of interferometers for dimensional change, by automatic micrometers for flow and by the use of an Olsen testing machine of 20,000 lb. capacity for compressive strength. The methods were in accordance with the conditions laid down by the Federal Specifications Board's requirement (No. 356) for dental amalgam alloys and the Bureau of Standards Technological Paper No. 157.

Copper amalgam obviously failed to meet the requirements of a specification which called for 65 - 70 per cent by weight of silver, but its dimensional

change was within the set limits of -0.04% to $+0.10\%$.

SCHARFENBERG (1948) was the first worker to publish data of any quantity on the early setting patterns of copper amalgam (i.e. compressive strength plotted against time). He found that at 1, 2, 3 and 24 hour intervals copper amalgam exhibited half the compressive strength of two silver amalgams selected for comparison.

His result for copper amalgam (5 brands) was 110.2 N/mm^2 at 24 hrs (the mean of 8 specimens), the corresponding result for silver amalgam (2 brands) was 258 N/mm^2 (the mean of 3 specimens). Clearly, not only was the number of specimens of copper amalgam and silver amalgam tested too few for valid conclusions to be drawn but it seems also unlikely that the 1, 2 and 3 hour specimens of copper amalgam were set.

He concluded that:

- "1. Copper amalgam does not set as rapidly as silver amalgam under any of the manipulative conditions investigated (i.e. controlled heating at 150°C and 170°C and heating over a Bunsen flame to 150°C).
2. The setting pattern of copper amalgam (i.e. compressive strength plotted against time) varies with the method used to heat the pellets.
3. Copper amalgam sets more rapidly as one approaches a minimum temperature to heat the pellets for trituration.
4. Setting intricacies exist in the copper

amalgams regardless of the manipulation of the pellets making it difficult, if not impossible to produce copper amalgam samples or restorations of identical characteristics."

Scharfenberg's comparative 24 hr. compressive strength results for copper and silver amalgams, however, do not compare with those of Taylor (1929) for the identical amalgams. Taylor (1929) had also observed that it was difficult to produce copper amalgam specimens of identical characteristics.

Scharfenberg also found that an increase in heat of preparation of copper amalgam increased the setting time (as defined by the mean compressive strength 60 minutes after mixing). The number of samples tested is too small for valid conclusions to be drawn. (15 at 150°C and 5 at 170°C).

For compressive strength tests he used smaller specimens than normal to simulate the size of restoration which would be found in the deciduous tooth. The specimens were 2.5 mm long and 2.5 mm in diameter and were made in a split die. A correction factor was used to make the results comparable to those of other workers. TAYLOR et al (1949) pointed out that 4 x 8 mm specimens gave 2 - 7 per cent higher compressive strength values than 6 x 12 mm specimens.

MATHEWSON (1970) obtained tensile strength data using the diametral compression test. His

results were as follows:

Type or Brand	No. of specimens	1 week strengths N/mm ²	
		—	s
Micro grain alloy silver	11	43.4	11.3
Silver-copper alloy 2.1: 2	10	31.9	8.8
Neosilbrin	9	18.4	3.5
Caulk's	10	16.6	4.8
Cupro Muc	13	10.4	2.2

after MATHEWSON (1970)

The method of heating the copper amalgam is not fully described. The value of 43.4 N/mm² for a modern silver amalgam seems low and the standard deviation for this amalgam, which should have been the most consistent performer in the group, seems large by current standards.

The diametral compression test or Brazilian test has been used for assessing the tensile strength of silver amalgam, (EDEN & WATERSTRAT, 1967, KORAN & ASGAR, 1967, BASKER & WILSON, 1971). MATHEWSON (1970) appears to be the only worker who has subjected copper amalgam to this test. According to BERENBAUM and BRODIE (1959) this test compares well with the conventional stretch tests for brittle materials which are anisotropic (e.g. concrete). The more brittle

the material the less the disc or cylinder is likely to be distorted before failure occurs and, according to PELTIER (1954), the test is valid provided the width of the contact area does not exceed one fifth of the specimen diameter.

DIMENSIONAL CHANGE ON SETTING

The dimensional change on setting of silver amalgam has always been characterised by an initial contraction before any subsequent change is revealed. This initial contraction is of the order of 0.04 per cent in amalgams containing 65 - 70 per cent silver and can exceed 0.14 per cent in those containing less silver, (SKINNER, 1946). It is not uncommon for high tin alloys to show a total contraction in 24 hours of 0.20 - 0.30 per cent, (TAYLOR, 1929, SHERMELDINE & SMITH, 1966).

Copper amalgam exhibits a setting contraction in the form of a simple exponential curve. TAYLOR (1929) gives results of 0.06, 0.015 and 0.048 per cent for the contraction at 24 hours of the three copper amalgams he examined. According to SKINNER (1936) the amount of contraction varies and, under certain circumstances, may be up to 0.3% at 24 hours. In 1946, as a result of work carried out by WORMINGTON (1939) under his direction this figure was reduced to 0.2% at 24 hours. Wormington heated twenty pellets of unspecified size for 4 minutes in a small test tube immersed in a paraffin bath, the temperature

of which could be regulated accurately. He triturated the amalgam manually, mulling in a fingerstall timing each operation. He did not control trituration pressure. He packed the amalgam into a 10 mm mould of unspecified diameter at 44N with a 3 mm round condenser point (10 thrusts per increment of unspecified size). The dimensional change was measured at room temperature (20 - 25°C) by interferometer over a 25 hr. period.

His results were as follows:

Temperature to which copper amalgam pellets were heated (°C)	Time at that temperature (min.)	Dimensional Change (%)
158	4	0.054
158	8	0.054
158	16	0.074
168	4	0.068
179	4	0.098
188	4	0.092
198	4	0.084
208	4	0.114
218	4	0.097
228	4	0.112
238	4	0.104

after WORMINGTON (1939)

He concluded that:

1. The trituration time did not affect the dimensional change.
2. The dimensional change always took the

form of a contraction which was reduced at the lower heating temperatures.

3. Higher heating temperatures caused a delay in the rate of contraction in the early setting period.
4. Within reasonable limits the time of heating has little effect on the dimensional change, but a prolonged time increases the delay period.

COGGAN (1933) in unpublished data obtained at the University of Michigan according to Federal Specification No. 356 obtained comparable results for dimensional change from 0.06 - 0.12% contraction at 24 hours.

LAVIS (1954) states also that copper amalgam contracts but produced no evidence to support this.

WORNER (1936) states that copper amalgams containing 61 - 72% mercury exhibit negligible contractions or small expansions with reasonable manipulative techniques but gives no evidence to support this and is the only worker to suggest that copper amalgam expands on setting.

Silver amalgam, after an initial contraction, shows a final expansion on setting. Thus it is important to define the time after the start of trituration at which measurement commences.

RANTANEN (1961), FUSAYAMA (1964) and JORGENSEN and HOLST (1964). If there was clear evidence that final

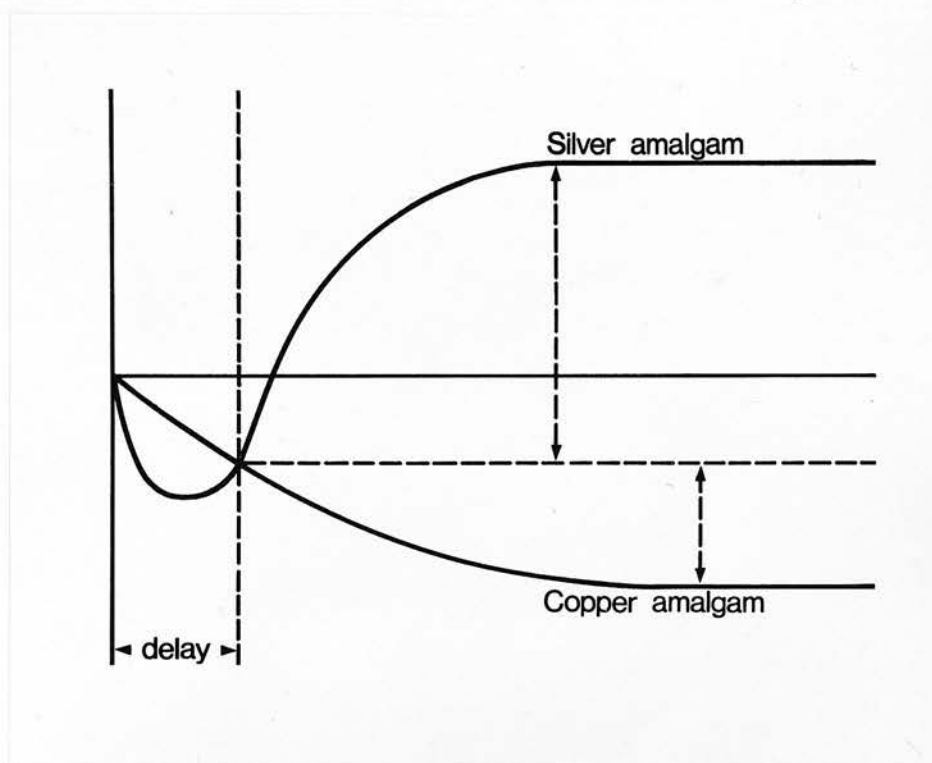


Fig. 4

expansion of the order normally exhibited by silver amalgams was desirable, a delay in commencement of measurement could increase final expansion of a silver amalgam and reduce the contraction of copper amalgam. (Fig. 4.) Thus opposite characteristics apparently alter results to support the use of the materials. Current specifications for the measurement of dimensional change of silver amalgams state that measurement should begin 15 minutes after the start of trituration. SMITH (1967) suggests that this time will be reduced to 5 minutes in later standards. He presents an example which shows that in the first few minutes after condensation of a silver amalgam the rate of contraction is rapid and that as much as 6 micrometres of an initial contraction were lost in an example as a result of a delay of 10 minutes in commencing measurement.

No details of the time at which measurement commenced are given by TAYLOR (1930) or WORMINGTON (1939) in their work in the dimensional change of copper amalgam.

TAYLOR (1929) tested the three copper amalgams for flow by applying a load of 250 Kg/cm^2 , 3 hours after amalgamation. All three gave values of 0.1%.

GRANATH (1961), investigating the hardness of silver and copper amalgams, prepared test specimens by using a machine which applied a constant packing pressure of 10 N/mm^2 . The copper amalgam was heated

by grinding the pellets flat on one side and placing them on a glass plate heated by a spirit flame. No heating temperature is specified.

The hardness test was carried out on an Alpha Durometer with a 2.5 mm diameter indenter. The vertical movement of the indenter could be followed without distortion, by means of a special linkage, on a dial gauge. The technique used for the test was to preload the specimen at 10 Kg by means of the indenter ball, then increase load to a predetermined total for 30 seconds and then reduce the load to 10 Kg. The difference between the readings taken before and after the application of total loads of 31.25 Kg and .25 Kg were recorded as the impression depth from which the hardness could be calculated. All specimens were prepared according to DIN and ISO standards for Brinell hardness.

According to the equation

$$HB = \frac{F}{D \times h}$$

where F = the load in kilogrammes force (Kgf)

D = the diameter of the ball in millimetres

h = the depth of the indentation in millimetres.

The Brinell Hardness (HB) was calculated from the data given by GRANATH (1961) as follows.

Amalgam	Alloy/mercury ratio	HB		residual mercury %
		F = 31.25	F = 25	
Silver fine grain preamalgamated	5: 5	61.27	59.1	40.1
Silver coarse grain	5: 8	58.6	53.0	50.4
Copper amalgam 1		88.5	83.8	65.3
Copper amalgam 2		86.5	83.8	63.8

Calculated from the data given by GRANATH 1961

The condensation technique was checked by grouping hardness values for each end of the specimens and by packing an additional set at a higher packing force. The mean values for elastic recovery after unloading to 10 Kg and calculated minimum impression depth for the appearance of elastic recovery are as follows:

	Amalgams			
	Silver	Silver	Copper	Copper
Total load 31.25 Kg.	5.2	5.0	8.7	8.6
Total load 25.00 Kg.	2.4	2.9	6.0	6.3
Calculated impression depth x .001 mm	45	49	22	17

after GRANATH (1961)

His conclusions were:

1. Copper amalgam has a statistically greater hardness than silver amalgam in

spite of a significantly higher mercury content.

2. Although the registration of the elastic recovery is inadequate for the determination of elastic properties, copper amalgam shows greater elasticity than silver amalgam.
3. That copper amalgam showed no tendency to flow.

It should be noted that the above results were obtained commencing the tests at the end of 24 hours and not after 3 hours as in A.D.A. Specification No. 1.

MATHEWSON (1970) carried out his flow tests according to the American Dental Association No. 1 apart from the fact that he used a mechanical amalgamation. His data are as follows:

Type or Brand	% flow <u>in</u> 24 hrs. Average of 4 specimens	Range %
Micro grain alloy silver	4.20	3.70 - 4.70
Silver copper alloy 2.1: 2	0.47	0.30 - 0.60
Neosilbrin copper amalgam	0.03	0.03 - 0.03
Caulk's copper amalgam	0.02	0.00 - 0.02
Cupro Muc copper amalgam	0.18	0.10 - 0.30

after MATHEWSON (1970)

These results were obtained by applying the 250 Kg. load to the specimen 3 hours after trituration commenced.

Many mechanical methods of specimen preparation have been described and undoubtedly those which produce consistent results and prepare specimens under conditions similar to clinical practice merit consideration, (BASKER & WILSON, 1969). SMITH (1967) stated, in effect, that lack of standardisation in previous amalgam studies resulted in generalisations which were doubtless true but could not be taken as an indication of what happened in clinical conditions. He assumed, however, on the slender basis of two examples, that satisfactory agreement could be obtained from data produced by workers in different laboratories using different apparatus!

In an effort to simulate conditions in the mouth numerous mechanical tests have been devised for dental materials. Many of these tests were suitable for a range of similar dental restorative materials but few have been standardised (SMITH, 1967). The commonest method of testing the ability of a material to withstand a maximum load is to apply a steadily increasing load in an axial direction to a cylinder of the material until failure becomes apparent. The nominal strength of the material is found by dividing the relevant maximum load by the area of cross-section of the cylindrical specimen.

TWEEDDALE (1964) implies that very little compression testing is done on metal since most metals are ductile. Compression instability and frictional restraint are given as the reasons for this.

30

Compression instability increases with the length of the specimen but is unlikely to occur unless the length to diameter ratio exceeds 3: 1. Frictional restraint takes place at the contact faces on the end of the cylinder when deformation occurs under the load and induces axial tension on the surface of the specimen. These limitations to compression testing of conventional metals and alloys do not apply to the same extent to copper amalgam for the following reasons:-

- (a) Copper amalgam is extremely brittle.
- (b) In the present study the specimen length to diameter ratio was 2: 1.
- (c) Compression tended to force the structure of the copper amalgam specimens closer together and crack propagation, which could occur due to porosity in the orthodox tensile test, was not a problem.
- (d) Frictional restraint at the contact faces increases with the diameter of the end of the specimen and while it obviously could not be avoided in the present study it was reduced to negligible proportions by the nature of the specimens.

A SUMMARY OF THE PHYSICAL PROPERTIES OF COPPER AMALGAM

COMPRESSIVE STRENGTH at 24 hr.

Number of
specimens

N/mm²

TAYLOR (1929)	263	
	303	
	331	3
SCHARFENBERG (1948)	110 mean	8

TENSILE STRENGTH at 7 days

MATHEWSON (1970)	10 - 18	32
------------------	---------	----

DIMENSIONAL CHANGE ON SETTING (PER CENT)

WORMINGTON (1939)	0.05 - 0.11	not stated
COGGAN (1933)	0.06 - 0.12	not stated
TAYLOR (1929)	0.048 - 0.06	3

BRINELL HARDNESS

HB

Calculated from the data given by

GRANATH (1961)	84 - 88	10
----------------	---------	----

The number of specimens of copper amalgam which have actually been subjected to mechanical tests is therefore surprisingly small in view of the fact that the material is still in use today.

ADAPTATION STUDIES

DEFINITIONS

MICROLEAKAGE STUDIES

These studies demonstrate whether or not fluid exchange takes place from the oral fluids to the tooth/filling interface of dental restorations. These studies have arisen due to the lack of a filling material which would adhere to the cavity walls.

One of the factors governing microleakage is the adaptability of the filling material.

ADAPTABILITY STUDIES

These studies demonstrate the degree to which filling materials may be approximated to the surface of the cavity wall. Two factors influence adaptability (1) The surface finish of the cavity wall and (2) the surface roughness of the restoration at the cavity wall.

ADAPTATION STUDIES

BALDWIN (1897) sought to improve the marginal seal of amalgam restorations by applying a thin mix of zinc oxyphosphate cement to the floor and walls of the cavity prior to condensation in the hope that the amalgam would adhere to the margins. He took care to ensure that the cement film was kept to a minimum and that the margins of the finished amalgam restoration were free of excess cement. He subjected extracted teeth having been restored by this technique to microleakage tests using ink and found that they were "watertight". KRAUS (1945) makes unsubstantiated claims for the clinical success of this restorative technique but in the light of present knowledge, however, the "stickiness" of the cement while setting is no indication of any adhesive properties it may possess when set. Therefore, as an attempt to obtain an adhesive restoration, the technique is unacceptable and, according to ALLAN (1957), lends itself to misuse. In a survey of dental restorations he examined sections of 12 extracted teeth which had been restored by the Baldwin technique and found that the cement had been completely removed from the cavity floors leaving an undesirable excess elsewhere.

The concept of "caulking" or sealing the space which exists between the cavity wall and the restorative material, however, is acceptable and is supported by the work described in the review of literature using copal varnish with silver amalgam

restorations. GOING and MASSLER (1961), BRANNSTROM and SOREMARK (1962) and BARBER et al. (1964).

Inlay technique also follows this concept although the preliminary function of the cement, in shear, is retention. NELSEN et al. (1952) give 44 micrometres as the average cement film thickness between the cavity wall and gold inlays. WING (1966) found spaces from 9 - 25 micrometres between the cavity wall and silver amalgams. It is important, therefore, to study the basic properties of adaptation of a restoration before the effects of any sealer can be evaluated.

JØRGENSEN (1965) states:-

"For amalgams good adaptability means good retention and diminished risk of penetration of saliva, food debris, bacteria, etc. In addition, good adaptability means a more perfect support of the filling margin, a factor which is undoubtedly of essential importance to the duration of this margin."

He studied the properties of adaptability of 10 silver amalgams and 3 copper amalgams by packing them, by different techniques, into a steel mould. The surface roughness of the mould walls and specimens was measured by means of a "Perth-O-Meter" Type V1 BC and graph recorder, Type R 120 n. The "Perth-O-Meter" is an instrument which is able to trace the surface profile by means of various diamond cone styli, in this case type HT 25/6 having a point radius of 10

54

micrometres was used. This stylus was able to measure depths not exceeding 25 micrometres at a load of 80 mg. which, Jørgensen states, was adequate for the greater number of test specimens. The graph recorder produced a trace of the surface profile magnified x 20 horizontally and x 1,000 vertically from which the arithmetic mean roughness (R_a) was calculated. In addition, an assessment of adaptability was made from the degree of reproduction on the freed specimen of lines or grooves of varying fineness engraved on the wall of the mould. Considerable care was taken to ensure that the surfaces produced were representative of a given combination of variables.

He produced data which indicated that the copper amalgams showed good adaptability. Poorer adaptability, as a result of a decrease in packing pressure was a characteristic of both silver and copper amalgams. Of the silver amalgams only one pre-amalgamated alloy gave results similar to those for copper amalgam for the same technique.

Amalgam Brand	Type	Ra Values	
		Packing 40 Kg. for 3 minutes standard	Pressure 15.6 Kg. for $\frac{1}{2}$ minute
New True Dentalloy	(Five grams silver)	1.2	2.3
Standalloy	(pre-amalgamated silver)	1.1	1.7
Argos	(pre-amalgamated silver)	0.6	2.3
Argos	(non-zinc, pre- amalgamated silver)	1.3	1.6
True Dentalloy	(zinc-free silver)	1.7	3.0
Cupro Muc	(copper amalgam)	0.6	1.5
Neo Silbrin	(copper amalgam)	0.6	1.3
Ash's Globe	(copper amalgam)	0.8	1.1
Polished steel	(mould surface)	0.03	

after JØRGENSEN (1965)

The test specimens were prepared by applying the above forces continuously to an upper moving piston against a lower fixed piston which formed part of the mould. The measurements were made when the specimen could be removed from the mould without damage and when repeated readings on the same profile gave the same results (approximately 24 hours). No details about the preparation of the copper amalgam are given.

HATT (1959), investigating the relationship of the amalgams to the cavity wall, used a similar instrument, the "Talysurf" to measure the surface roughness of silver amalgam (New True Dentalloy)

packed against a control surface of glass. He expresses the roughness as "the average size of the spaces between the amalgam specimen and the cavity wall" and gives the following data from 194 specimens:

Mould of glass and steel to simulate proximal box	Average size of spaces (micrometres)	
	between amalgam specimen and cavity wall	along cavo surface angle
Cylindrical condenser area = 1.80 mm^2 , 1.5 mm dia.	1.67	3.55
Trapezoidal condenser area = 2.32 mm^2	1.54	1.93
Mechanical condenser Dentatus area = 1.80 mm^2 , 15 mm dia.	0.63	0.96

after HATT (1959)

The ideal force for hand packing, using a cylindrical condenser 1.5 mm diameter, was found to be 35.58 N (19.32 N/mm^2). The force applied to the Dentatus mechanical condenser with a cylindrical tip of 1.5 mm diameter was 2.22 N.

McHUGH (1955) packed 40 amalgams into acrylic dies having cylindrical cavities 8 mm deep and 4 mm diameter. The main feature of these dies is that a fine thread is cut in the cavity wall to provide a standard test of adaptation. He used a "fairly large-grained" amalgam alloy prepared by a controlled

technique and condensed the amalgam by hand and by vibrator. Three days after packing the dies were sectioned and polished. The extent to which each thread section on each specimen was filled was examined and a simple score allotted accordingly.

He found:

- (1) Vibrator packing produces better adaptation than hand packing.
- (2) The condensation of small increments produced better results than the Bergendal technique (i.e. where packing is started by condensing a small amount of amalgam using a small vibrator point then the cavity is filled to excess, the amalgam covered with a celluloid strip and a large vibrator point applied).

No packing pressures are given, only a reference to heavy hand pressure.

Thus, having examined the adaptability of various amalgams (i.e. the degree to which various amalgams can be adapted) and the adaptation (i.e. the state to which the adaptability was carried under a given set of circumstances) without reference to the dental tissues, a technique which would permit an examination of the state of affairs in vivo was sought.

MICROLEAKAGE STUDIES

NELSEN et al (1952) attempted such an investigation. Restorations inserted in extracted teeth and in cavities blown in glass (no exact details are given) were immersed in ice cold water for 30 seconds. The margins were examined under a stereomicroscope as the specimen warmed up in the fingers. They were able to demonstrate that:

1. There was a fluid exchange between the tooth substance and various restorations caused by changes in temperature.

They concluded that:

- i. The fluid exchange was caused in part by the difference in coefficients of thermal expansion of tooth substance, restorative material and the fluid between the restoration and the cavity.
- ii. The fluid exchange might explain recurrent caries.

They were also able to measure temperature changes in three teeth, one in each patient, by means of a 36 gauge copper-constantan thermocouple placed on the pulpal wall and held in place by a self-curing acrylic resin restoration. The lowest temperature recorded was 9°C caused by drinking water at 4°C and the highest was 52°C caused by drinking coffee at 60°C , giving a change of 43°C . No measurements were attempted under metal restorations.

GOING et al. (1960a) investigated the marginal penetration of restorations placed in 316 freshly extracted teeth by immersion for 24 hours in a solution of radio-active sodium iodide (I^{131} ; 25 - 50 microcuries/cm³; pH 6.0 - 5.2) in distilled water to which was added 0.2 cm³ crystal violet dye (0.025% aqueous solution). The technique by which the cavities were cut, the teeth stored, sectioned and autoradiographs prepared was standardised. An unspecified number of restorations which had been in place for at least 12 months prior to extraction were also examined for comparison. There was no control over cavity preparation or preparation of the restorative material in this group. The permeability of freshly cut dentine was also assessed by immersing the unfilled cavities in 47 teeth in the tracer solution. This served as a control experiment and also established that 24 hr. immersion was sufficient to achieve maximum penetration. The dye used in vivo on two teeth in different patients substantiated this finding.

The conclusions were:

- (1) Freshly cut dentine in unfilled cavity preparations was equally permeable to crystal violet dye and I^{131} .
- (2) All restorations showed some degree of penetration by I^{131} .
- (3) Under the conditions of the experiment,

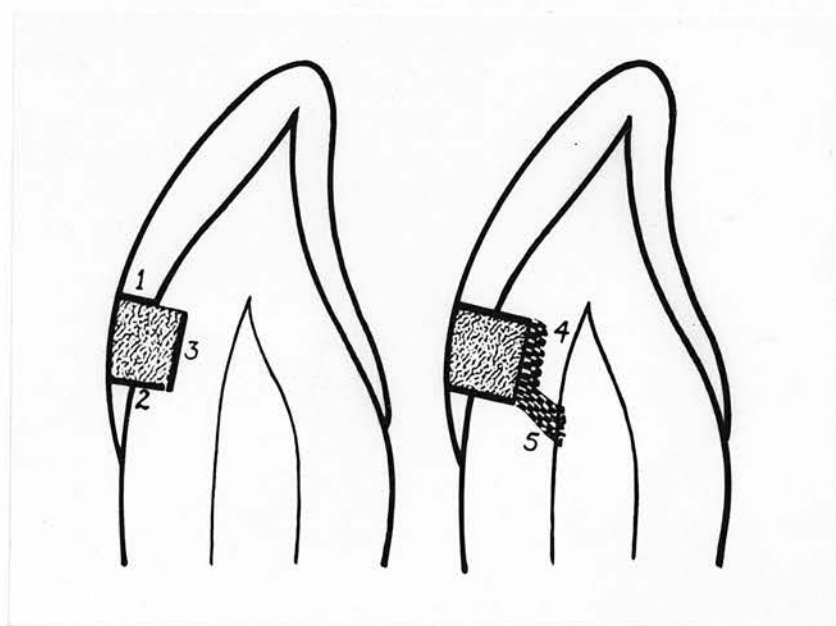


Fig. 5

Index of penetration depth for Fig. 5a

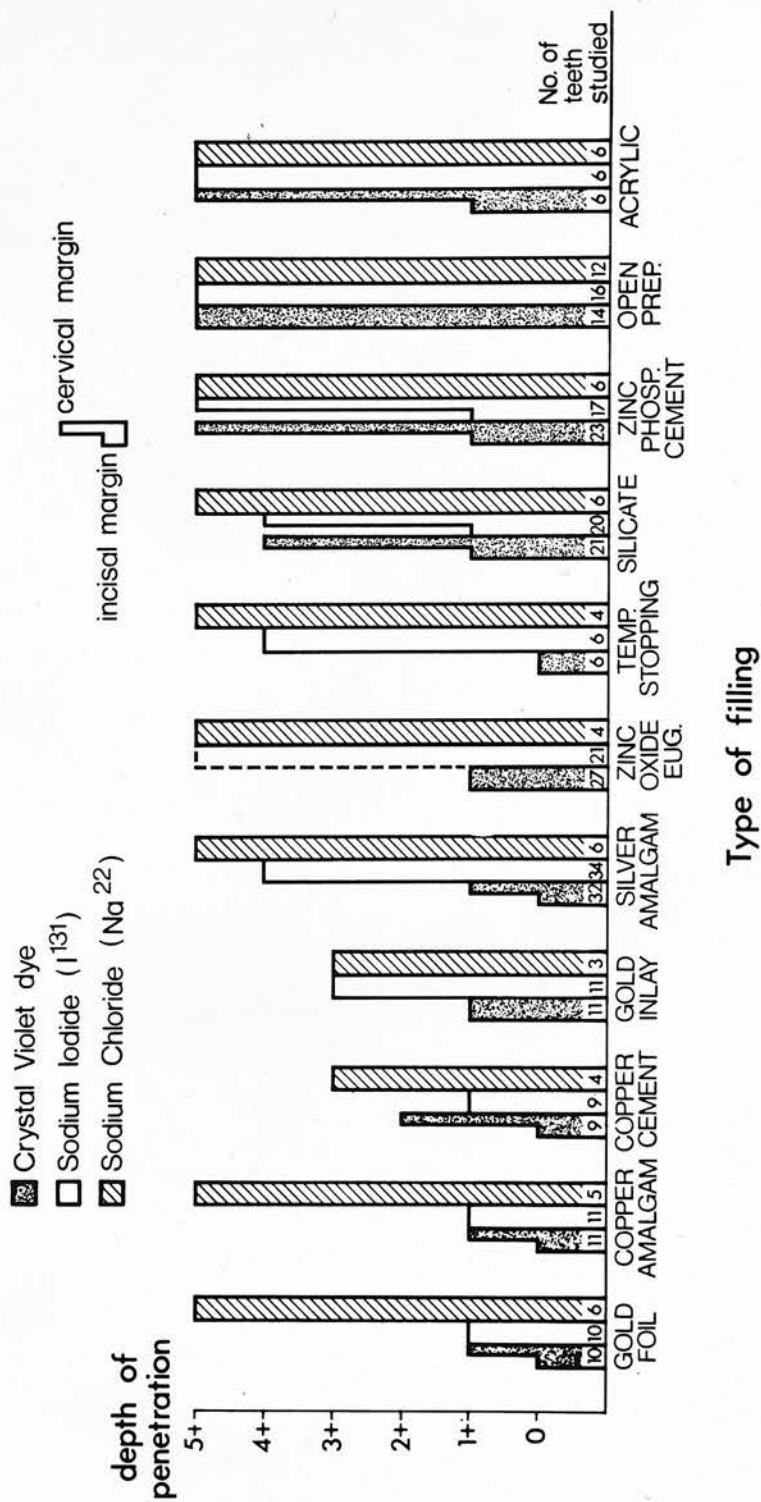


Fig. 5a

after GOING et al. 1960 (amb)

gold foil, copper amalgam and red copper cement allowed minimum penetration of the isotope and the dye (Fig. 5) while for the other materials penetration varied but, in general, was greater than for these three materials. Silver amalgam showed considerable penetration round the margins and into its substance.

- (4) Intact enamel margins were an important factor in the reduction of microleakage.
- (5) Silver amalgam restorations and silicate restorations over a year old allowed less marginal penetration than freshly placed restorations. No explanation for this was given.

However, in a contemporaneous investigation in vitro of the marginal penetration of dental restorations using the radio-active isotopes S^{35} , P^{32} , Na^{22} , Rb^{86} and Ca^{45} as tracers, GOING et al. (1960b) were able to demonstrate the penetration of Na^{22} to the pulp chamber around gold foil and copper amalgam restorations. The experimental conditions were identical to those of the study mentioned previously.

Their basic conclusions were:

- (1) That all the dental restorations

allowed penetrations at the margins by one or more of the tracers.

- (2) "The ionic charge and chemical activity of the ion, as well as the physical and chemical nature of the restorative material, influence the depth of marginal penetration of the isotope".
- (3) "In general, the metallic restorations were superior to the non-metallic restorations under the conditions of this experiment."
- (4) "A single order of marginal permeability could not be established, since the order of penetration varied somewhat with different isotopes."

GOING and MASSLER (1961) on experiments on 234 extracted teeth with silver amalgam restorations found that copal resin, polystyrene-ethylcellulose and calcium hydroxide liners were effective in preventing marginal penetration of radioactive ions into dentine and pulp tissue. The teeth were not subjected to variations in temperature. BRANNSTROM and SOREMARK (1962) restored 32 teeth with silver amalgam in vivo using contralateral pairs one of which was lined, the other unlined in each pair. They showed that the penetration of the radioactive isotope Na^{22} (as physiological NaCl) was reduced by the

application of a calcium hydroxide varnish on all the exposed dentine. Exposure of a group of teeth with lined cavities to temperature variations of 36 centigrade degrees increased the degree of penetration considerably and doubts were expressed about the ability to reduce marginal penetration in vivo. BARBER et al. (1964) flooded the cavity walls as well as the cavity floor with copal varnish immediately before condensing the amalgam restoration and obtained similar reductions in penetration using toluidine blue dye and S^{35} as a radioactive tracer.

GRANATH (1967) compared the marginal leakage in vitro of two silver amalgams, one a preamalgamated medium-cut alloy, mercury-alloy ratio of 5: 5 and the other a course cut alloy, mercury-alloy ratio 7: 5, and a copper amalgam (5 specimens each).

The amalgams formed part of the base of epoxy resin cups which were filled with isotonic saline and placed in isotonic saline in which the sodium was in the form of radioactive isotope Na^{22} . Microleakage was measured by sampling from within the test cups at specified intervals and a comparison made between cups subjected to temperature cycling before the experiment and those subjected to the same cycle during the experiment.

He found that there were microleakage differences which he considered were consistent with their dimensional change on setting, up to a period of 30 days. Copper amalgam showed much greater leakage

than silver amalgams. Temperature cycling had no effect if carried out on the test cups before the experiment and produced inconsistent results when carried out towards the end of the microleakage tests. Granath suggests two experimental errors:

- (1) The leakage test carried out at room temperature.
- (2) No account taken of the water absorption of the epoxy resin.

He also draws attention to the fact that in this experiment the Na^{22} ions were shown to pass in both directions through microleakage pathways.

MCDONALD & PHILLIPS (1950) placed 60 restorations in vivo using a silver amalgam which contracted 0.28 - 0.34% on setting as measured by well established bench tests. Clinical examination of 30 of these restorations over a period of 3 years and the remainder for shorter periods failed to produce evidence of contraction or recurrent caries while in some cases caries were seen to destroy adjacent teeth. These results were, however, obtained without the aid of radiographs. (BARR and GRESHAM (1950) have pointed out that radiographs are an essential aid in any examination to detect the presence of carious lesions in teeth. They examined existing restorations in 162 young adults and found that from a total number of defects discovered by complimentary radiographic and visual clinical inspection, 38.8% were

disclosed clinically and 84.2% radiographically.)

SWARTZ and PHILLIPS (1962) made use of two amalgams in vitro to check on the significance of dimensional change in marginal penetration. One amalgam contracted 0.35% on setting while the other expanded 0.25%. Results using Ca^{45} as a tracer from a minimum of 8 restorations of each amalgam demonstrated the same degree of penetration for both amalgams. Thus, doubts were cast on the long held and seemingly logical theory that a setting expansion would be a major factor in the sealing of the cavity margins.

The A.D.A. specification No. 1, which designated a setting expansion of zero to 0.20%, has now been altered (1971) to allow a dimensional change on setting of $\pm 0.20\%$. *

From the extensive literature on the use of radioactive isotopes at the margins of dental restorations two factors emerge which appear to bring about a reduction in penetration under silver amalgam restorations:

- (1) The age of the restoration. A freshly placed restoration allows increased marginal penetration by radioactive isotopes when compared with a similar restoration which has been in place for 1 - 2 weeks.

* Measurement commencing 5 min. after start of trituration

- (2) The use of a cavity varnish or silver which acts in the manner of a "caulking agent" on the floor and walls of the cavity.

WING (1966) was sceptical about the comparability of results obtained by bench adaptation tests and the use of radioactive isotopes in any quantitative sense. In addition, spherical particle amalgam alloys began to appear on the market with higher early strengths and easier condensation claimed as advantages. He too placed silver amalgam restorations made from conventional (lathe cut) alloy and spherical particle alloy in cavities cut in freshly extracted molar and premolar teeth. He varied (1) the condensation technique (good and poor) and (2) applied a copal varnish liner (thick and thin) to some of the cavities. Three storage media at 37°C were used for the teeth for periods ranging from 1 day to three months.

- (1) Distilled water.
- (2) Saliva.
- (3) 10% sodium sulphide.

The teeth were sectioned and polished taking care to avoid flow in the amalgam, and then etched. The space between the restoration and the cavity wall was then measured under the microscope using a filar eyepiece. As a check on the accuracy of this method, certain of these prepared sections were embedded in acrylic resin under vacuum to ensure that the interface

was filled below the surface. The specimen was resectioned and polished arranging for a new surface level 0.5 mm below the original, thereby preventing any distortion by polishing.

He found:

- (1) That the space between the amalgam and the tooth is real, as shown by the presence of acrylic resin in the space, and not an artifact produced during the preparation of the specimen.
- (2) With good condensation, typical width of interface space would be:

	Micrometres		
	cavo-surface	lateral walls	pulpal wall or floor
(a) Lathe cut alloy	19	7	7
(b) Spherical particle	25	16	23

Poor condensation increased the spaces significantly with conventional alloys but made little difference to the already larger spaces produced by the spherical particle alloys.

- (3) The application of a lining of copal ether varnish to the cavity floor or pulpal wall increased the width of the space between the amalgam and cavity wall by approximately 8 micrometres.

Thick application increased the dimension by 20 micrometers. Some areas were seen to be free of varnish particularly on the lateral walls of the cavity and some lateral and corner spaces of 80 - 100 micrometres were seen.

"

BERGVALL & BRANNSTROM (1971) used the scanning electron microscope (S.E.M.) to measure the space between the cavity walls and composite resin restorations. They did not regard the method on its own as being reliable. They obtained results from 34 restorations placed in extracted premolar teeth and confirmed them with the aid of a light microscope and replica techniques. They found spaces of usually about 9 micrometres but the spaces varied considerably between 2 and 20 micrometres at the cavity floor. The spaces on the lateral walls were narrower varying from less than 1 micrometre up to 10 micrometres. An attempt to measure the amount of microleakage was made by GRANATH (1970). The technique used was similar to the methods of KNAPPWOST (1951) and PICKARD and GAYFORD (1965) who used air pressure to expose marginal leakage in restored teeth and made attempts to measure the space which would allow the leakages. Both attempts were based on theoretical rather than practical considerations. For example, PICKARD and GAYFORD (1965) did not take into consideration that there would be lateral

communication of leakage pathways at the interface.

Thus the validity of the concept of "total effective leakage area" is in question.

GRANATH (1970) produced sophisticated apparatus from a development of the above ideas for the quantitative comparison of microleakage in vitro. The air which passed through the margins of the test restoration was also passed through a capillary tube, the mouth of which was immersed in a liquid of low surface tension. The amount of air passed through the test piece was calculated from the volume and frequency of the bubbles collected and represented the leakage due to the available leakage area. The accuracy of the result depends on:

- (1) The accuracy of the flow meter and differential manometer used to measure the flow and pressure of the compressed air.
- (2) The maintenance of a steady pressure, slightly above atmospheric, on the distal side of the test piece.
- (3) Constant temperature round the test body ($37.0 - 37.1^{\circ}\text{C}$). This partly depended on the temperature of the compressed air being equal to that of the test body. (37°C)

The apparatus showed a high degree of accuracy (0.15% mean error) in calibration tests and was suitable for the purpose for which it was designed.

SUMMARY

JØRGENSEN (1965) standardised packing procedure and compared the adaptability of 13 dental amalgams. Copper amalgams (3) exhibited the highest degree of adaptability shared by one silver amalgam of the preamalgamated variety. McHUGH (1955) and HATT (1959) compared condensation techniques using a single type of silver amalgam and found that the condensation technique was an important factor in adaptation and that the use of a mechanical condensing instrument produced better adaptation than hand condensation.

NELSEN et al. (1952) demonstrated that there was fluid exchange at the margins of dental restorations and measured temperature range experienced by the natural teeth (9 - 52°C).

GOING et al. (1960a) and (1960b) used radioactive isotopes as tracers and the technique of autoradiography to study microleakage in restorations. They found that all restorations leak with both one or other of the tracers. GOING and MASSLER (1961), using the same technique, found that liners prevented microleakage. BRANNSTROM and SOREMARK (1962) found that use of a cavity varnish reduced microleakage but expressed doubts about the situation in vivo. BARBER et al. (1964) used a dye in addition to a tracer and found a reduction in microleakage with the use of copal varnish. GRANATH (1967) used isotonic

solutions with a radioactive tracer to measure fluid exchange in vitro but his results were inconclusive.

WING (1966) measured the size of the inter-space between cavity wall and amalgam restoration and found spaces of up to 100 micrometres. He compared the interspaces resulting from the use of spherical particle alloys with those of conventional alloys.

BERGVALL and BRANNSTROM (1971) found spaces of 1 - 20 micrometres between composite resin fittings and the cavity walls using S.E.M.

McDONALD and PHILLIPS (1950) found in a clinical survey of 30 restorations over a period of 3 years that an amalgam having a setting contraction (0.34 per cent) showed no increase in secondary caries. There was no control group, however, in this study. SWARTZ and PHILLIPS (1962) performed studies in vitro using two amalgams, one with a setting contraction and one with a setting expansion, and found no difference between the two. Thus the evidence available suggests that a dimensional change of approximately 0.30 per cent is not significant in relation to microleakage or the incidence of secondary caries.

GRANATH (1970) has developed a technique for measuring microleakage using compressed air.



EFFECTS ON THE DENTAL PULP

The aim in conserving a tooth is to restore form and function and whenever possible to ensure continued vitality of the dentine-pulp unit.

There are many factors in the operative field which influence the health of the dental pulp. Much has been written about the effects on the dental pulp of restorative materials placed directly against the cut surface of the dentine on the pulpal floor. There have been many studies of the trauma produced by certain methods of cavity preparation. It has been shown that comparable results in such investigations are dependent on two conditions:

- (1) Uniformity of thickness of dentine between cavity floor or pulpal wall and the pulpal tissue, referred to in current literature as the remaining dentine (R.D.). A hint of this was given by GURLEY and VAN HUYSEN (1937) in dog teeth and is well substantiated by STANDLEY and SWERDLOW (1959) for human teeth.
- (2) The ability to distinguish between the chemical effects of the restorative material per se and the effects of microleakage. There is no evidence that this has been possible.

MANLEY (1936), cut an unstated number of cavities in the teeth of dogs without causing observable damage to the pulp and, although he was aware that the cutting procedure alone would cause a stimulation of secondary dentine (KRONFELD, 1933), he was able to produce different pulpal reactions to different restorations. "No appreciable reaction" resulted from a restoration of zinc oxide/eugenol cement while "more serious" reactions resulted from the use of silicate and copper oxy-phosphate cements. Silver amalgam gave rise to a "well formed barrier" and "a considerable amount of secondary dentine". MANLEY made no mention of cavity depth. GURLEY and VAN HUYSEN (1937) substantiated Manley's results in an unstated number of experiments in dogs' teeth but found that red copper cement gave different reactions depending on the amount of dentine remaining between the restoration and the pulp. In their experiments the amount of dentine varied from 0.1 mm to 1 mm.

They concluded that:

"(1) Filling of cavities with the cements or temporary stopping is followed by changes in the underlying pulp and dentine histology.

(2) Destructive changes are evident when the cavity and filling lie close to the pulp; but secondary dentine is deposited when they are placed at a distance from the pulp. These phenomena may take place in the same tooth.

- (3) Reaction of the pulp may be prevented by filling or lining cavities with a mixture of zinc oxide and eugenol."

Present day pulp studies would appear to owe a great deal to the original work of MANLEY (1936) and GURLEY and VAN HUYSEN (1937).

In the absence of reports in the literature of the direct effect of copper amalgam on the dental pulp, some believed that it behaved like a mummifying agent causing slow devitalisation of the pulp. DICKSON and RICKERT (1933) showed that metallic copper and copper amalgam caused a much more violent reaction than any of the other dental restorative materials, including silver amalgam in rabbit soft tissue implant studies. EFFINGER (1954 and 1957) and MITCHELL (1959) obtained similar results.

Discolouration of the tooth substance surrounding the restoration of copper amalgam is well established and a green stain is characteristic. Manufacturers of certain brands claim to have overcome this disadvantage.

Attempts to detect mercury from silver amalgam restorations in pulp tissue using radioactive mercury Hg^{203} by FRYKHOLM and ODEBLAD (1955) produced positive but unreliable conclusions although the presence of mercury and metal ions in the dentine under silver amalgam restorations has been ascertained by MASSLER and BARBER (1953), SOREMARK et al. (1968).

These workers also found that a cavity liner protecting cut dentinal tubules effectively prevented the ingress of metal ions. This is also advocated by SWERDLOW and STANLEY (1962).

Experiments on the response of the human dental pulp to silver amalgam show that the effects are mild, reversible but surprisingly indistinct, considering the long and widespread use of this material. The effects of cavity preparation, remaining dentine thickness, microleakage, lining materials, the toxic effects of the material per se and the products of corrosion of the material have made investigations difficult to compare.

SWERDLOW and STANLEY (1962) claim that the problem is unresolved and the findings of more recent workers, GRANATH (1969) tend to be hypothetical.

SUMMARY

The effects of various restorative materials on the dental pulp was studied by MANLEY (1936) and GURLEY and VAN HUYSEN (1937) who placed restorations in the teeth of dogs. They found that continued vitality of the dentine pulp unit depends on the remaining dentine thickness (R.D.) and the effect of the material placed against the cut surface.

This has been substantiated by STANLEY and SWERDLOW (1959) for human teeth.

Although soft tissue implants show a modest reaction to copper amalgam, there is no evidence to show that copper amalgam has an adverse effect on the dental pulp.

Green staining of the tooth substance is a characteristic of certain brands of copper amalgam.

The detection of mercury in the dentine underlying silver amalgam restorations has been confirmed by MASSLER and BARBER (1953) and SOREMARK et al. (1968). The presence of mercury in the pulp from the same origin is probable but not confirmed. Cavity liners are effective in preventing mercury from entering the dentinal tubules.

Results from experiments designed to evaluate the effect of silver amalgam on the dental pulp are obscure.

OLIGODYNAMIC ACTION

MILLER (1890) was the first to report on the persistent antiseptic action of copper amalgam.

NÄGELI (1893) observed that spirogyra organisms were killed not only when kept in aqueous solutions in metal vessels but also by exposure to water that had been kept in metal vessels. HOGEBROOM (1923) exposed a culture of oral bacteria to restorative materials on culture plates. He found that copper amalgam inhibited growth but gold had no effect. HUSBAND (1928 and 1932) demonstrated that a plain copper wire has no antiseptic qualities when placed in infected agar, but the wire exhibited marked germicidal power when coated with copper amalgam prepared by immersing it in mercuric chloride (HgCl_2). He also suggests a method for obtaining copper amalgam for the sterilisation of root canals as an aqueous solution, but he provides no clinical evidence that his method did not cause periapical irritation.

FRASER (1929) demonstrated the germicidal action of copper amalgam by applying small cubes of the amalgam to pour plate cultures. For her early studies she used Bacillus coli and later exploited the alkalinity of Bacillus proteus cultures. Measurements of the area of inhibition were made after 48 hours and after 1 week using B. proteus. On approximately every eighth day the specimens were placed on new pour plates. This was continued until no inhibition was obtained on at least two successive transfers. The

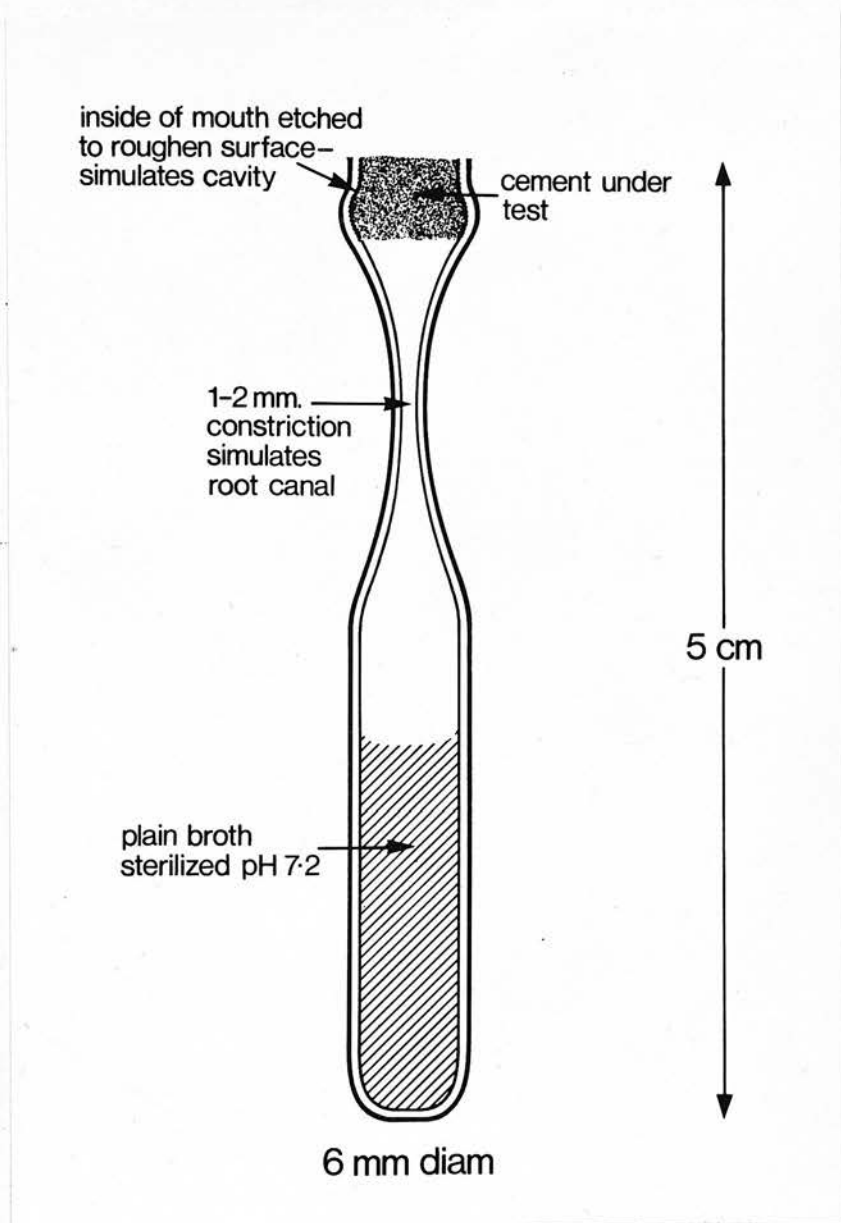


Fig. 6, after Fraser (1929).

diameter of the area of inhibitions of growth of bacteria increased from 7 mm. to 22 mm. on the fourth transfer, then decreased to 10 mm. and remained the same over the course of 50 successive transfers. The cubes of copper amalgam lost 49% of their original weight after being plated 50 times in $2\frac{1}{2}$ years.

FRASER also performed permeability tests using test tubes with a narrow section half way down their length to represent the cavity and root canal. Plain broth at pH 7.2 was placed in the lower portion below the narrow section and sterilised. The restorative material was placed at the entrance to the narrow section as a seal. This area had been etched to simulate conditions in a tooth cavity. The culture medium was placed in the open top portion covering the restorative material. (Fig. 6) Her results were inconclusive and temperature variation during the course of the experiment is given as the source of error.

SHEPPARD (1935) used gold and silver amalgam restorations from freshly extracted teeth and cylinders of pure gold, silver and copper to investigate the antibacterial properties of metal restorations and their components. Micro-organisms were obtained from tooth scrapings, pure cultures of Lactobacillus acidophilus, Staphylococcus aureus, a yeast and several strains of viridans streptococci isolated from human mouths. For culture media dextrose veal-infusion agar made up to ten times the specified

concentration was used as a base for the special media. Saliva agar was made by adding 10 cm.³ of this base to 90 cm.³ of dilute hydrochloric acid. The metals copper and mercury were found to be strongly bacteriocidal. Copper amalgam was not included in this study. Pure gold and silver possessed no bacteriocidal properties. Most gold inlays exhibited growth in plate cultures of L. acidophilus, streptococci and mixtures of mouth organisms, and in peptone water suspensions of L. acidophilus, but often appeared to enhance the growth of staphylococcus and yeasts. The amalgam restorations were found to have variable effects with no correlation to recurrent caries in the tooth of origin.

McCUE et al. (1951) found silicate cement to be the only other dental material to possess bacteriocidal power superior to that of copper amalgam. These workers admit to a limited number of platings and their results for acrylic resins are not substantiated by other workers.

TURKHEIM (1953) confirmed the prolonged bacteriocidal effect of copper amalgam and found it to be the most powerful of the dental materials in his study. He found no oligodynamic effect with silver amalgam alloy powder. When mercury was introduced, however, the freshly mixed amalgam produced some effect. It was more toxic to Staph. aureus than to Escherichia coli. Monilia albicans and Pseudomonas pyocyanea were not affected. Eight strains of L. acidophilus

odontolyticus were tested because they had shown preliminary sensitivity to silver amalgam. After 33 transfers of the amalgam over the course of several weeks, six of the strains were unaffected and it was evident that the silver amalgam gradually lost its bacteriocidal effect. Cadmium was found to be more potent as a bacteriocide than silver. (Cadmium is found in certain copper amalgams).

SMITHERS and PARFITT (1955) tested the following dental restorative materials for their effect on acid production by oral bacteria:-

1. Cast gold
2. Silver amalgam
3. Copper amalgam
4. Silicate cement
5. Self-polymerising resin
6. Zinc oxyphosphate cement
7. Copper cement
8. A proprietary brand of zinc oxide and eugenol

Cylinders with an exposed surface area of 200 sq. mm were attached to the inside of the screw-cap of 7 cm³ bottles containing 4.5 cm³ stimulated saliva and a 5 per cent concentrate of calcium triphosphate and dextrose solution. The pH of the saliva was measured before and after incubation for four hours at 37°C. Two bottles were used as controls and each material was tested six times. Results showed that

copper amalgam, the zinc oxide eugenol derivative and silver amalgam showed significant inhibitory powers.

Copper amalgam demonstrated the highest initial germicidal power completely inhibiting acid production and although diminution of this inhibitory power followed, a constant level was reached after 12 - 16 hours. Silver amalgams and the zinc oxide eugenol derivative showed a more rapid loss of inhibitory powers. Adequate checks were carried out and results from four other brands of silver amalgam showed no variation from that obtained in the original study.

They also found that the exposed surface area of copper amalgam had to exceed 220 mm^2 per 4.5 cm^3 saliva before any persistent effect was obtained in a static environment.

MATHEWSON (1970) tested 5 amalgams for their oligodynamic effect. These included proprietary brands of copper amalgam, one silver amalgam and a mixture of unstated proportions of silver amalgam and copper amalgam. The test micro-organisms used were L. acidophilus, a streptococcus, E. coli and a Neisseria species. The culture medium was "Bacto-Mueller-Hinton (Difco)" with blood, but E. coli was grown without blood. The cultures were maintained in a 10% carbon dioxide atmosphere at 37°C and zones of inhibition were measured after 12 hours.

<u>Amalgam</u>	Zone of inhibition in mm. observed with the stated organism			
	<u>E. coli</u>	<u>Neisseria</u>	<u>Strept.</u>	<u>L. acidophilus</u>
Neosilbrin copper	6.25	3.5	5.5	3.75
Caulk's copper	9.5	3.0	7.0	4.0
Cupro copper	5.25	12.25	8.0	NIL
Silver (Hg/alloy 7.5)	NIL	7.5	NIL	NIL
Silver/copper	NIL	12.00	NIL	NIL

after MATHEWSON (1970)

MATHEWSON stated that he related these tests to the optimum time for growth as for antibiotic sensitivity and that results could be affected after that time due to a reduction in the availability of nutrient materials or to an overgrowth of the medium by the organisms.

SUMMARY

The oligodynamic action of copper amalgam was first reported by MILLER (1890).

FRASER (1929) demonstrated the persistent germicidal action of copper amalgam, demonstrated a loss in weight of approximately 50 per cent in $2\frac{1}{2}$ years in the copper amalgam discs used in the study. Fraser attempted permeability tests using copper amalgam but results were inconclusive.

SHEPPARD (1935) found that copper and mercury were strongly bacteriocidal. McCUE et al. (1951) found silicate cement possessed bacteriocidal power superior to that of copper amalgam. TURKHEIM (1953) confirmed the prolonged bacteriocidal effect of copper amalgam. He found no such effect from silver amalgam alloy powder but when it was combined with mercury there was some bacteriostatic action which gradually deteriorated.

SMITHERS and PARFITT (1955) also found that copper amalgam was strongly bacteriocidal and found the size of the surface area of the copper amalgam specimen required to produce effective inhibition of acid production in a given volume of saliva.

MATHEWSON (1970) found that copper amalgam had superior bacteriostatic properties to those of silver amalgam.

MICROLEAKAGE STUDIES USING BACTERIA

MORTENSEN et al. (1965) repeated the work of FRASER (1929) with some modifications. The aim of the study was to develop a method for studying the penetration of bacteria around the margins of restorations in human teeth. Instead of placing the restorative materials in glass tubes, the extracted teeth containing the test restorations were sealed into plastic tubes with a commercial epoxy resin. Thus the tooth formed a barrier between a medium that was specifically infected on the coronal side and the same medium, maintained sterile, on the apical side. Serratia marcescens was used as an indicator organism and adequate tests and controls were carried out to ensure that contamination could only take place via the tooth-restoration interface. Growth of the test organism in the medium in the apical side was therefore an indication of micro-leakage. The pulpal wall or floor was removed from the cavity and access canals made so that no restrictions were placed on bacteria that managed to pass through the margins. Restorations incorporating (i) silver amalgam (ii) silicate and (iii) zinc phosphate cement were tested in this system.

The silver amalgams were incubated at 37°C for 72 hours, chilled until the infected medium on the coronal side of the tooth reached a temperature of +4°C to -6°C and then re-incubated at 37°C. If no leakage was apparent as indicated by the development

of turbidity in the medium below the apices in 24 hrs., the specimen was returned to 37°C and the chilling cycle repeated followed by a further 24 hrs. incubation after which a final check on turbidity was made.

Results for silver amalgam restorations were as follows.

Total number of silver amalgam restorations	Number yielding no micro-leakage	Number showing microleakage before thermal cycling	Number showing microleakage after thermal cycling
51	23	6	22

after MORTENSEN et al. (1965)

Thus under these experimental conditions, 55% of the silver amalgam restorations allowed bacteria (0.5 - 1.0 micrometres in size) to pass between the margin and the cavity wall. Thermal cycling, especially with the silver amalgams in this study, was shown to be a significant factor and substantiates the findings of NELSEN et al. (1952).

Serratia marcescens was chosen as a test organism because of its size, ease of identification and its successful use in the hands of other workers. This organism, however, is not generally found in the natural environment of the teeth. Mortensen et al. do not appear to have controlled the variation in cavity shape and size. While there is no evidence to show that this is a significant factor in micro-leakage studies of this type, it should have been considered.

In a previous study BROWN et al. (1962) used

Lactobacillus arabinosus. This organism requires niacin for growth. They were able to establish that when the growth of this organism was dependent on diffusion of niacin from within the tooth (i.e. the apical medium in vitro) in-vitro carious lesions were produced that simulated natural caries.

If, however, in vitro conditions were arranged so that niacin was only obtainable from an external source (i.e. the coronal medium, with sterile water substituted for the apical medium), the lesions produced did not simulate natural caries.

It would appear that the problem is mainly one of availability of the nutrient factors while the source, despite the diffusion theory, might be of secondary importance. These teeth were maintained in vitro in a static environment not subject to temperature variations.

ELLIS and BROWN (1967) used a different approach:-

- (1) "To determine the feasibility of a specific in vitro technique for studying caries at the interface of the dental restoration and the tooth.
- (2) To employ this method to evaluate lined and unlined amalgam restorations in preventing or inhibiting in vitro caries round restorations."

28 unerupted human third molar teeth were used in this study. Two cavities of similar size were cut in occlusal pits of each tooth and silver amalgams placed, one was lined with cavity varnish (MIZZY handi-liner), the other unlined. Variation in cavity size was evident from photographs presented. After sterilisation of the tube containing the tooth a niacin-deficient medium was placed on the coronal side of the tooth and a solution containing niacin (100 micrograms/cm.³) was placed on the apical side. The medium on the coronal side was seeded with Lactobacillus arabinosus 17/5 (ATCC 8014) and the tubes were incubated at 37°C for periods of 12 and 16 weeks. The medium on the coronal side was changed and the teeth rinsed with sterile water at weekly intervals. pH measurements were used as an index of growth in the media. Results showed that the unlined restorations were more likely to produce caries than the lined restorations. This is in agreement with the radioactive isotope microleakage studies of GOING and MASSLER (1961), BRANNSTROM and SOREMARK (1962) and BARBER et al. (1964).

ATTO et al. (1970) carried out a similar experiment in vitro to compare the ability of unlined copper amalgams and silver amalgams to inhibit dental caries. 23 teeth were used, each having one copper amalgam and one silver amalgam restoration placed on the occlusal surface. Carious lesions, indistinguishable from natural caries, occurred round 9 of the 23

silver amalgam restorations and around 18 of the 23 copper amalgams. The cavity size and shape were again variable. The greater number of carious lesions round copper amalgam restorations was attributed to the setting contraction and disintegration of the material.

However, no details are given about the properties of the amalgams used in this study, their preparation or the packing technique. The statement made by the authors that "silver amalgams expand slightly on hardening" is true but does not mean that all silver amalgams exhibit a final setting expansion.

While the result of this study is of interest because it is contrary to what would be expected, the technique incorporates too many variables that have not been taken into account. In addition, the reasons given for the results are based on wrong assumptions.

SUMMARY

The aim of these studies was to study the penetration of bacteria around the margins of human teeth. MORTENSEN et al. (1965) demonstrated that thermal cycling (0° - 37° approximately) significantly increased the penetration of bacteria between the restoration and the cavity wall.

BROWN et al. (1962) demonstrated that Lactobacillus arabinosus, which requires niacin for growth, could obtain it from within the tooth substance by diffusion and under these conditions the resultant carious lesion simulated natural caries.

ELLIS and BROWN (1967) developed this experiment to examine the effects of cavity liners. Two small silver amalgam restorations were placed in each tooth, one lined, the other unlined. They showed that the unlined restorations were more likely to produce caries.

ATTO et al. (1970) carried out a similar experiment to compare the ability of copper amalgams and silver amalgams to inhibit caries. One restoration of each type was placed in each tooth and it was found that more carious lesions were related to the copper amalgams than to the silver amalgams. This result was attributed to the setting contraction and disintegration of the copper amalgam under these conditions.

CLINICAL STUDIES

GRANATH & HAKANSSON-HOLMA (1961) carried out a comparative clinical study of copper and silver amalgam on 63 pre-school age children. The cavities selected for the copper amalgam restorations had suffered more carious destruction than those selected for silver amalgam. Follow-up examinations were carried out when the average age of the restoration was two years.

Marginal defects were charted for each surface. The authors found different types of defect occurred in the same surface. Thus few conclusions could be drawn but in general copper amalgam restorations showed fewer marginal deficiencies than silver amalgams.

Secondary caries frequency was significantly higher in the silver amalgam group (6.9%) compared with the copper amalgam group (1.1%) but the authors point out that secondary caries often occurred in connection with other defects.

Good adaptability and a virtual absence of flow are suggested as principal reasons for the superior clinical performance of copper amalgam in this study. It was not possible to decide whether the lower occurrence of secondary caries was due to the bacteriostatic effect of the copper amalgam or its better marginal adaptation.

It is clear that the number of variables in this study was considerable. Defects which

occurred on cavity margins as a result of faulty cavity preparation could not necessarily be attributed to the properties of the amalgam. Evidence of such defects might have been obliterated by secondary caries. The use of a clinical cavity form makes any form of measurement difficult. Thus identical cavities of similar depth on contra-lateral teeth should be used in studies of this type.

MERCURY HAZARDS

TOVERUD (1929) and BORINSKY (1931) have shown that there was a higher degree of mercury excretion via the urine and faeces from patients with copper amalgams than from patients with silver amalgams 3 days after the insertion of the restorations.

FRYKHOLM (1951) has confirmed BORINSKY'S work but shows that the expected risk from mercury poisoning is not as serious as at first imagined.

In a preliminary report STEWART and STRADLING (1971) compared the mercury concentrations* in three different dental surgery situations. They found that in a surgery of 49.5m^3 , near the point of preparations of one copper amalgam pellet, the concentration exceeded $770\text{ ug}/\text{m}^3$ which was the maximum reading of the meter and, 15 minutes later, that the average concentration in the room with no exhaust ventilation had dropped to $140\text{ ug}/\text{m}^3$.

* Mercury concentration Meter Type E3472 (Columbia Industrial Developments, Ltd., Bath Road, Slough, Bucks.)

An extractor fan reduced this to 75 ug/m^3 in 5 minutes. The Department of Employment in the United Kingdom (1968) set the present maximum threshold limit value for mercury vapour in air during continuous occupational exposure at 100 ug/m^3 , and it is intended to reduce this to 50 ug/m^3 in future legislation.

FINAL SUMMARY

In a clinical study GRANATH and HAKANSSON-HOLMA (1961) demonstrated that copper amalgam gave better clinical results when compared with a silver amalgam in deciduous molars. In a group of 63 pre-school children, there was a significant difference in the secondary caries incidence in favour of the copper amalgam restorations. Good marginal adaptability and lack of flow are given as reasons for its superiority. Although this is the only clinical study in the literature to provide statistical data, the number of variables is considerable.

Marginal adaptability in vitro for 3 copper amalgams was demonstrated by JORGENSEN (1965) to be better than that of 5 silver amalgams. The adaptability of one silver amalgam, made from a preamalgamated alloy, was almost as good as that of the copper amalgams used in the study. In the literature this was the only study of adaptability which included copper amalgams.

Preliminary microleakage studies, GOING et al. (1960), using I^{131} suggested that restorations of copper amalgam (also gold foil and red copper cement) showed minimal marginal penetration compared with other filling materials. However, later studies using several radioactive isotopes separately, showed that all restorations including copper amalgam allowed marginal penetration to one or other. Silver amalgam exhibited considerable marginal penetration which decreased with age. No clear quantitative information has emerged from these studies.

TAYLOR (1929), COGGAN (1933) and WORMINGTON (1939) produced comparable data on the dimensional change on setting for copper amalgam which shows that it contracts 0.05 - 0.12 per cent. SCHARFENBERG (1948) demonstrated a reduction in crushing strength with an increase in the heat of preparation. WORMINGTON (1939) showed also that there was an increase in the setting contraction with an increase in the melting temperature. The number of specimens which were tested in the above studies was too few for valid conclusions to be drawn.

The minimum temperature required to prepare copper amalgam for dental use is $150 - 160^{\circ}\text{C}$ (SCHARFENBERG, 1948 and HUTCHESON, 1969). A temperature of preparation is not mentioned in any of the instructions for use with copper amalgams currently produced.

TAYLOR (1929) found that the crushing strength values (263, 303 and 331 N/mm^2) at 24 hrs. for three copper amalgams were similar to those values he obtained for amalgams made from alloys which contained over 65 per cent by weight of silver. SCHARFENBERG (1948) produced data for 1 hour compressive strength of copper amalgam using minimal heat (150°C) for preparation. It seems highly probable that an imperfect melt resulted or that the amalgam had not set in the time. He also used a non-standard specimen size in this test.

TAYLOR (1929) gives flow values of 0.1 per cent for three copper amalgams. GRANATH (1961) demonstrated elastic properties in copper amalgam and found that it did not flow.

In hardness tests GRANATH (1961) showed that copper "amalgam" was significantly harder than the silver amalgam tested, despite a significantly higher mercury content than the silver amalgam.

The metallurgy of copper "amalgam" is obscure but may, to some extent, account for its erratic behaviour when subjected to certain laboratory tests. It is not, to be accurate, an amalgam but is an electron or inter-metallic compound of copper and mercury. Chemically, such compounds are of closely controlled composition, ususally involving small, simple ratios of the combining elements where the laws of valency are not obeyed. Physically, such compounds are brittle but of variable hardness. The speed of formation of copper amalgam, which is slow, corresponds to the crystallisation or "ageing" of the compound, (CHAO & COSTA, 1968). This contrasts with the dental silver amalgam particle/matrix system where a comparatively fast interaction takes place between mercury and the silver/tin alloy particles.

The fact that copper and its derivatives are known to be toxic to plant life and bacteria has given rise to the belief that copper "amalgam" would be endowed with more powerful bacteriocidal properties than silver amalgam. FRASER (1929) and TURKHEIM (1953) confirmed this by placing small pieces of copper "amalgam" and silver amalgam on four plate cultures and comparing the areas of inhibition of growth. This is, perhaps, a naive approach to the problem but there is no other evidence to show that the mercury or copper in copper amalgam are not both active constituents.

70

In one artificial caries study, in vitro, ATTO et al. (1970) used extracted teeth with copper amalgam and silver amalgam restorations. They found copper amalgam to be less effective than silver amalgam in preventing the occurrence of lesions simulating dental caries.

Implantation studies in the soft-tissues of animals show that severe reactions result from the use of copper amalgam (DICKSON & RICKERT, 1933, EFFINGER, 1957, MITCHELL, 1959) but there is no evidence to show that these findings apply to the dentine-pulp unit.

In any study of copper amalgam, therefore, the questions at issue would appear to be:

- (1) Do copper amalgam restorations retain well compared with silver amalgam restorations under exactly the same conditions of use and if so, how is this achieved?
- (2) Microleakage has been demonstrated at the cavity margins of copper amalgam restorations. This has been ascribed to the setting contraction of the material. The significance of the setting contraction is obscure, but the evidence available suggests that the microleakage is minimal and the adaptability good compared with silver amalgam restorations.

What mechanism, therefore, contributes to the reduction in secondary caries is shown by

the clinical studies of GRANATH &
HAKANSSON-HOLMA (1961) when ATTO et al.
(1970)* found an increased caries
incidence related to copper amalgam
restorations in vitro?

CHAPTER I
SOME MECHANICAL PROPERTIES
OF
COPPER AMALGAM

* P.67.

METALLURGICAL ANALYSIS

Four different copper amalgams (containing 10, 20, 30, and 40% copper) were subjected to compression tests. The results are given in the following table:

Amalgam	Yield	Ultimate
10% Cu	20.3	29.3
20% Cu	22.2	27.7
30% Cu	20.1	27.2
40% Cu	27.2	30.2

CHAPTER 1

SOME MECHANICAL PROPERTIES

OF

COPPER AMALGAM

Brand	Approximate 1-2%
Amalgam (New Formula)	Castable
Cavalier	
Copper Alloy	Castable, Indur and Tin
New Formula	Castable, Silver and Tin

METALLURGICAL ANALYSIS

Amalgams are used for dental restorations. The composition of amalgams varies from about 10% to 40% copper. The mechanical properties of amalgams are affected by the composition and the method of preparation. The following table gives the mechanical properties of four different copper amalgams.

PRELIMINARY STUDIES OF COPPER AMALGAM

1. CHEMICAL ANALYSIS

Four brands of copper amalgam (2 samples of each) were subjected to spectrochemical analysis and found to contain the following amounts of mercury and copper:-

Brand	Hg%	Cu%
Ash's (New Formula)	80.2	20.3
Caulk's	72.2	27.7
Cupro Muc	70.1	27.2
Neo Silbrin	67.2	30.8

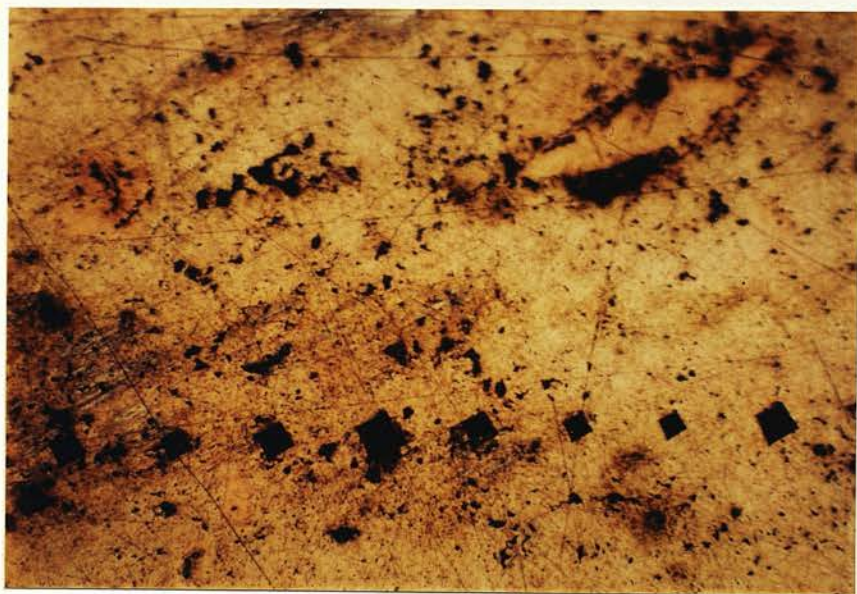
The same brands were then examined by X-ray fluorescence* and found to contain three additional metals as follows:-

Brand	Approximately 1-2%
Ash's (New Formula)	Cadmium
Caulk's	-
Cupro Muc	Cadmium, Indium and Tin
Neo Silbrin	Cadmium, Silver and Tin

2. METALLURGICAL EXAMINATION

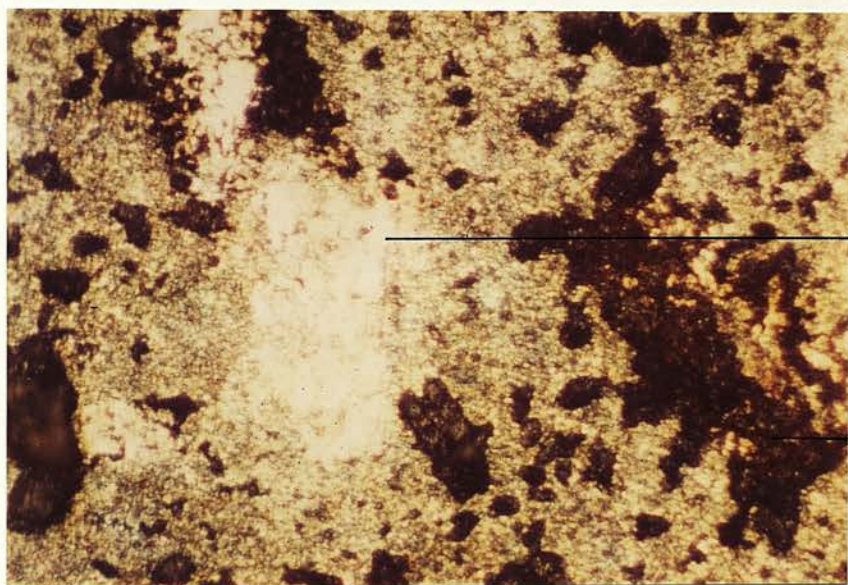
Specimens of the above brands of copper amalgam were prepared for examination by normal polishing techniques and finished using diamond pastes. They were examined under incident light by optical microscope. No etchant was used as it was found that the

*Phillips Manual X-ray Spectrometer Type P.W. 1540



x 250.

Fig. I. Ia



copper
rich
area.

porosity

x500

Fig. I. Ib

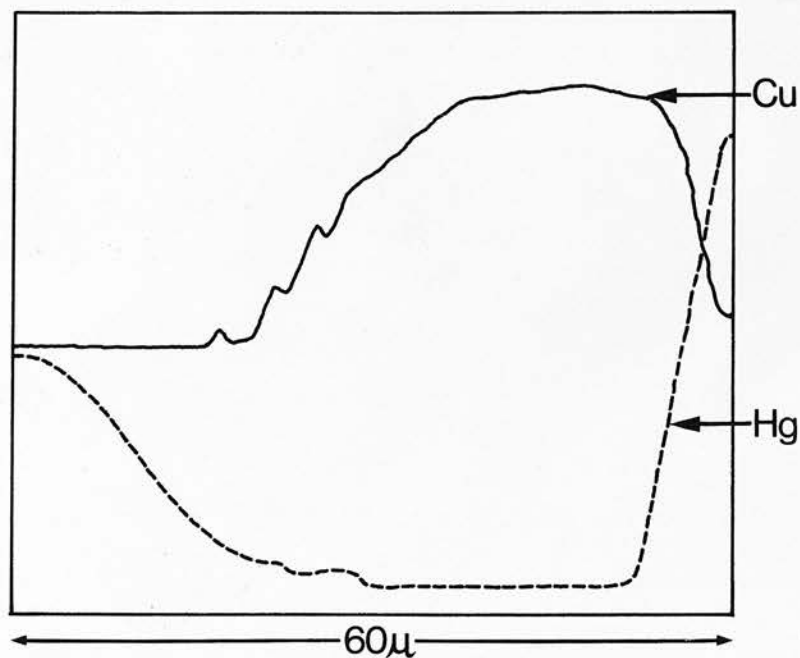
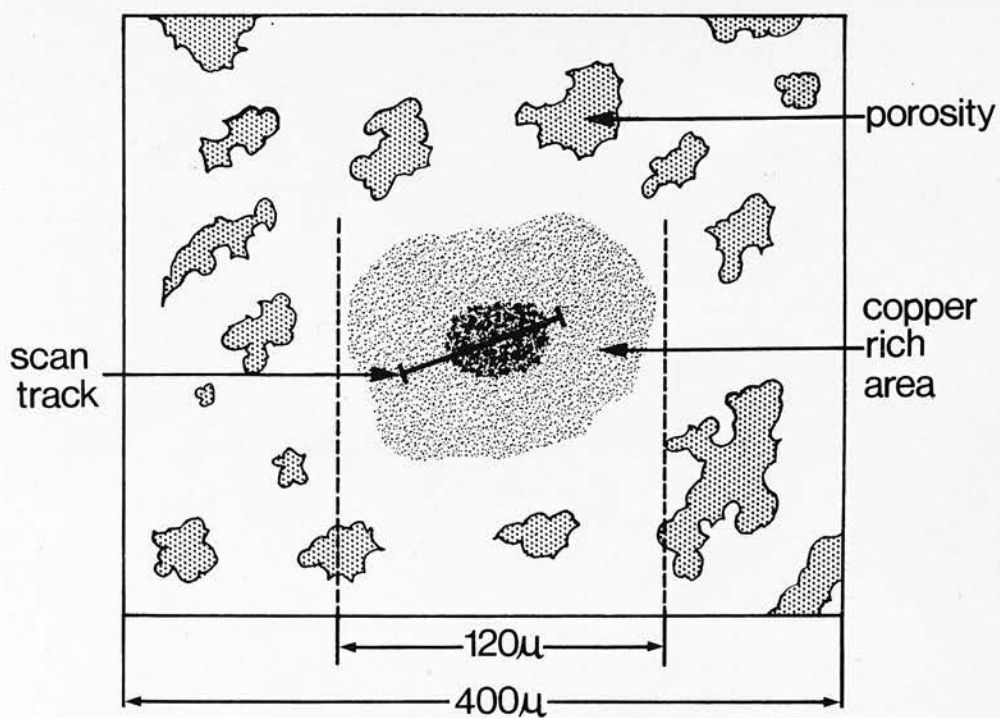


Fig. I-2

(Upper). Diagram of optical field as seen in the electron microscope.

(Lower). Plot of x-ray fluorescence intensity above the metal surface for copper and mercury from scan track shown in upper diagram.

two phases present in all the amalgams contrasted sufficiently as a result of oxidation. One phase, distributed at random appeared to be copper rich, the other mercury rich (Fig. 1.1a and b).

Porosity of varying degrees was present in all the amalgams examined (2 of each brand).

A microhardness test tried on the sample in Fig. 1.1a shows the wide variation in hardness which would appear to be due to porosity and lack of homogeneity. Results varied from 59 - 97 (BHN)^x.

3. ELECTRON PROBE ANALYSIS

Two specimens of copper amalgam were subjected to electron probe analysis*. Standards for copper and mercury were not available for quantitative analysis but examination confirmed the existence of areas 20-100 micrometres in diameter almost entirely composed of copper. The second phase was almost entirely composed of mercury (Fig. 1.2).

The larger copper rich areas exhibited relief polishing which showed that this phase was the harder of the two. There was a considerable amount of porosity in both samples and the instrument detected copper richness on one side of the pores in the specimens. This was assumed to be the detritus from the harder copper rich areas which had gathered there as a result of the polishing technique.

* Cambridge Instruments - "Microscan".

^xBHN = Brinell Hardness.

THE MECHANICAL PROPERTIES OF COPPER AMALGAM

DIMENSIONAL CHANGE ON SETTING

MATERIALS and METHOD

Six brands of copper amalgam were used in this study, three of which are no longer available.

PREPARATION OF COPPER AMALGAM SPECIMENS

The pellets of copper amalgam were placed in a 10 cm³ flask surrounding the bulb of a 220°C mercury thermometer which rested in a constant position in the base of the flask. The flask was heated by a 30W electric heating element* controlled by a 7500 ohm variable resistance which was adjusted to give a temperature rise of 5°C per minute. The heating apparatus provided full insulation for the flask (Fig. 1.3).

The hot pellets were then shaken into a normal amalgam mortar at room temperature and any free mercury poured off. They were reduced by hand to an even paste using a mortar and pestle. Measurement of dimensional change began 15 minutes after start of trituration.

Excess mercury was removed by squeezing the mix in a dental gauze and further excess was removed during condensation.

Cylindrical specimens 10 mm long by 5 mm in diameter were made by packing the amalgam into a steel mould which had been stored at 37°C.

The specimens were packed by hand using a smooth

*"Isomantle" by Isopad, Ltd., Boreham Wood, Herts.

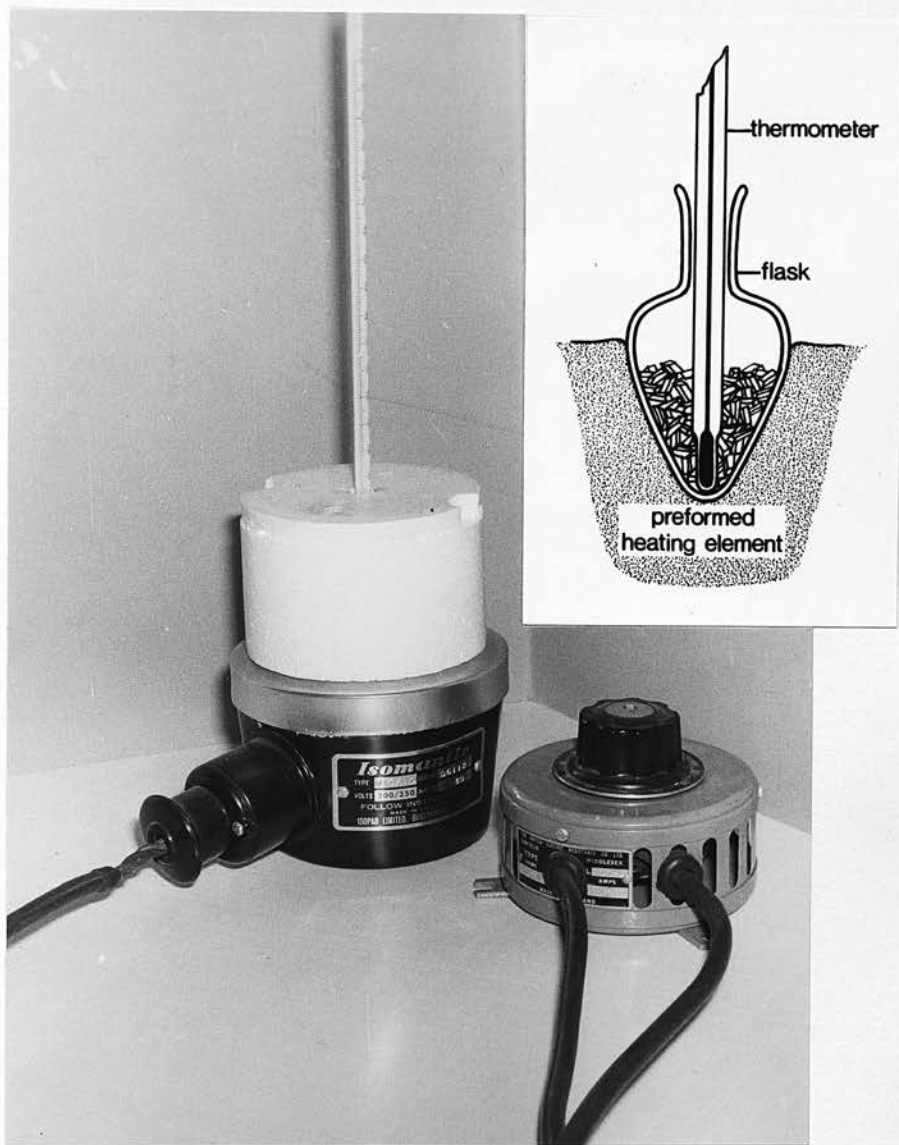


Fig. 1.3

Apparatus used to heat the copper amalgam pellets.



Fig. I.4

The "Mitronic" micro comparator used for measuring dimensional change on setting.

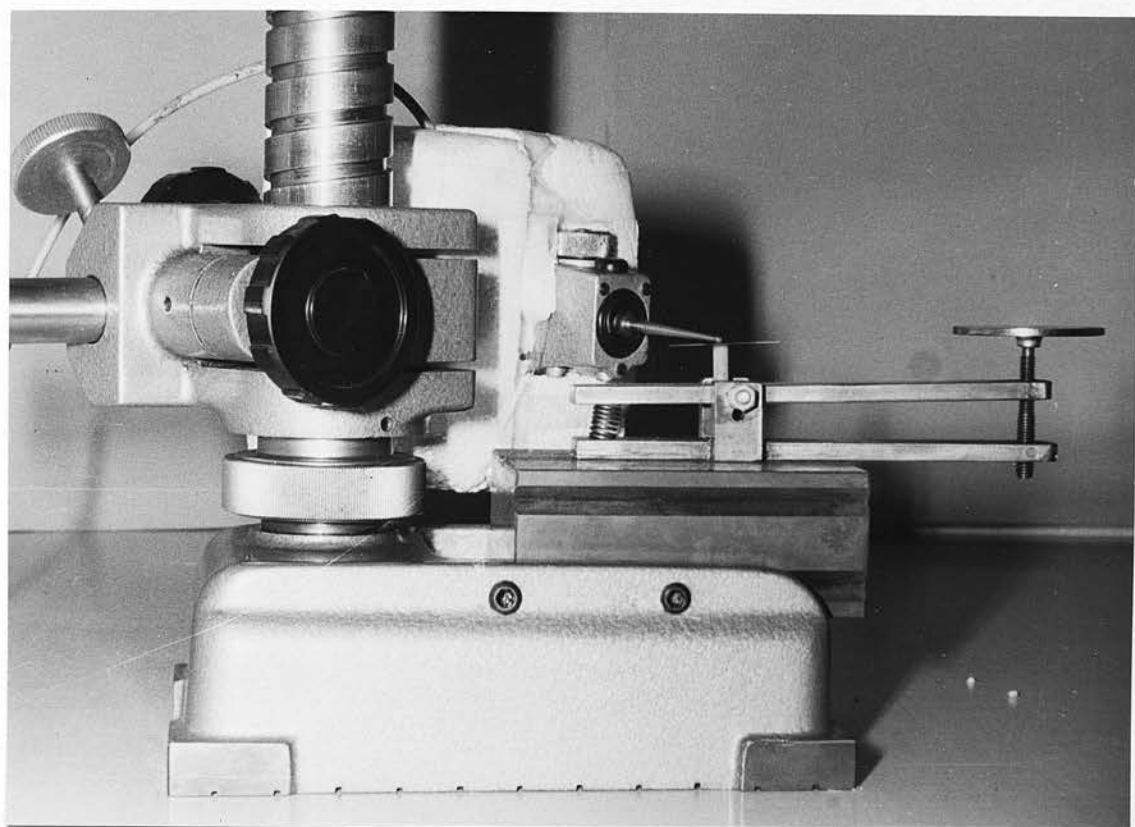


Fig. I.5

Close up of the "Mitronic" stand showing the use of the differential screw (right) to adjust the stylus height. (See text page 62).

cylindrical condenser of 2 mm diameter. Approximately 100 thrusts of between 35 and 44 N (8 and 10 lb.) were used to pack each specimen. The force of each thrust of the condenser was measured by placing the steel mould on a device which indicated when each condenser thrust was within the required force range and recorded the total number of condenser thrusts. Owing to the amount of mercury expressed from the copper amalgam during packing, the number of increments required to fill the mould was almost double that required for a silver amalgam of the same size.

APPARATUS

The apparatus for measuring dimensional change comprised three parts.

- (a) A measuring head with stand
- (b) An indicator unit which could be switched to any one of five scales. This was connected to the measuring head by flexible cable.
- (c) A pen recorder linked to the indicator unit.

Working Principle

A transducer in the measuring head converted the movement of the side acting stylus into an electrical signal in the form of a voltage change. This change was amplified by a transistorised circuit in the indicator unit and used to operate a voltmeter.

The "Mitronic" micro-comparator by Rank Taylor Hobson, Leicester (Fig. 1.4).

The transducer comprised an inductance bridge of which the four arms were an integral part of the measuring head. This bridge was energised by an oscillator contained in the indicator unit. As the moving part of the transducer was directly coupled to the stylus, mechanical linkages and lost motion were eliminated and minute movements of the stylus were measurable.

The measuring head had an adjustment which allowed the stylus to be preloaded by deflecting it. The stylus preload was proportional to the length of scale selected. It was defined as the load required to bring the pointer from full scale deflection to zero (i.e. the point of balance of the inductance bridge).

A stylus preload of 3 gm. at zero was used in this study to maintain the stylus tip in contact with the specimen. A glass coverslip (0.1 gm) was placed on top of the specimen to prevent the stylus tip sinking into the surface.

Preliminary studies revealed that the variation in length of the specimens exceeded 60 micrometres. The use of clamps supplied with the apparatus to adjust the stylus height from specimen to specimen introduced errors of up to 5 micrometres which were thought to be due to the effects of creep in the special clamping screws. A device to obviate use of the clamps was made in the form of a simple lever (Fig. 1.5). The specimen was placed close to the fulcrum at one end of the lever and a differential screw with a wide range of adjustment fitted to the other end. Variations in specimen length exceeding

twice full scale deflection were thus accommodated and control was sufficiently precise to enable zero to be set on the indicator unit by this means alone. The measuring head with sample was maintained at a temperature of $36 - 38^{\circ}\text{C}$ in an incubator, the door of which was adapted to provide screened access for the hands only. The temperature in the area of the specimen was checked by a thermistor in a bridge circuit. This was superimposed on the dimensional change graph on the pen recorder by an intermittent pulse carried via a relay to the pen recorder.

The steel die for specimen production was kept in the incubator with the apparatus for measuring dimensional change. It was also used as a slip gauge to test the accuracy of the apparatus before the study began and at random during the study.

The accuracy of the instrument was checked by measuring the differences in thickness of glass cover-slips (130-150 micrometres thick). The readings on the 30 micrometer scale of the "Mitronic", which was used in this study corresponded to within 1 micrometer of a metric bench micrometer screwgauge calibrated to read direct in 1/500's of a millimetre.

RESULTS

Dimensional Change on Setting at 24 hrs. - per cent

Measurement began 15 minutes after start of trituration.

Cylindrical specimens 10 x 5 mm.

COPPER AMALGAM - overall results

Temperature of preparation	No. of specimens (n)	\bar{x}	s
160°C	45	0.123 contraction	0.042
200°C	16	0.136 contraction	0.046

Comparison of dimensional change of COPPER AMALGAM by brands.

Temperature of preparation - 160°C

	(n)	\bar{x}	range
Ash's New Formula	11	0.095 contraction	0.075-0.120
Cupro Muc	8	0.128 contraction	0.070-0.170
Neo Silbrin	8	0.099 contraction	0.080-0.120
Caulk's	6	0.200 contraction	0.160-0.220

Temperature of preparation - 200°C

Ash's New Formula	4	0.132 contraction	0.070-0.160
Cupro Muc	4	0.118 contraction	0.100-0.145
Neo Silbrin	4	0.113 contraction	0.100-0.130
Caulk's	4	0.181 contraction	0.110-0.260

SILVER AMALGAM - room temperature

New True Dentalloy	6	0.071 expansion	0.060-0.085
Amalcap	6	0.068 contraction	0.070-0.100

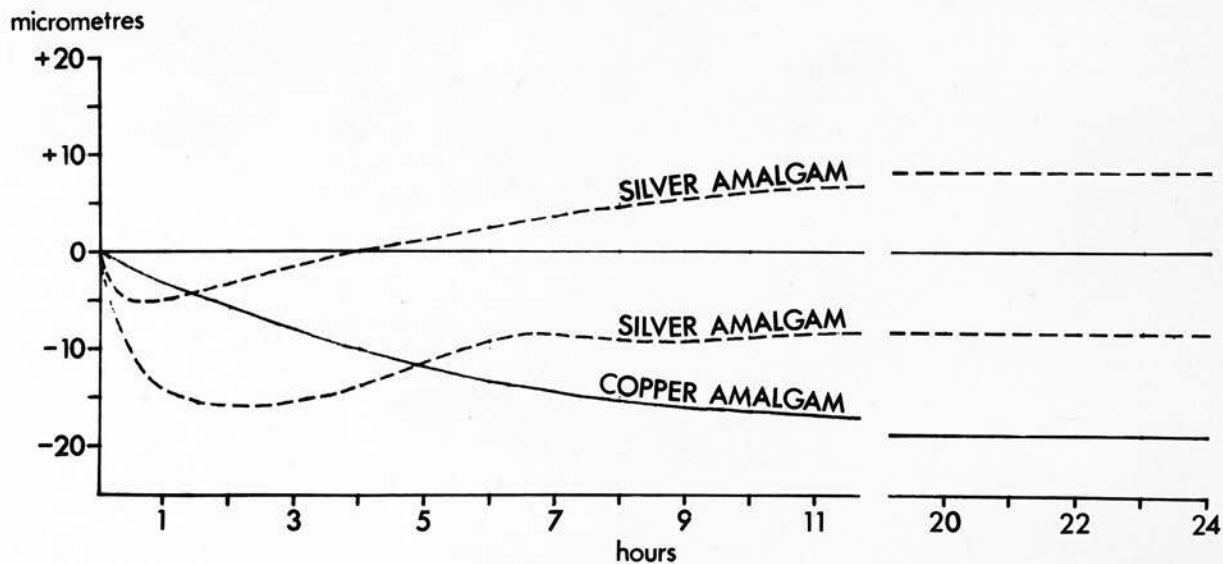


Fig. I.6

Typical examples of dimensional change plotted against time for copper amalgam and two currently used dental silver amalgams.

(Adapted from the pen recorder traces)

DISCUSSION - Dimensional Change

All the copper amalgams tested exhibited a setting contraction in the form of a simple exponential curve as shown in Fig. 1.6. This characteristic was the most consistent of those tested. All the results were less than the 0.300% contraction which was found to be of no clinical significance by McDONALD & PHILLIPS (1950) and even allowing for the altered time of commencement of measurement, most of the results would comply with the $\pm 0.20\%$ in A.D.A. Spec. No. 1.

There was no apparent difference in setting contraction between the two temperatures of preparation (160°C and 200°C).

COMPRESSIVE STRENGTH and TENSILE STRENGTH

The copper amalgam was prepared as before. Six cylindrical specimens 10 x 5 mm were made from the one heated amount. As some specimens produced by extrusion immediately after packing tended to bend, a split mould was used in this study so that the cylinders of copper amalgam could set without distortion.

The specimens were crushed in a Hounsfield Tensometer using the 1016.05 Kg (1 ton) beam for compressive strength. The Tensile Strength was carried out by subjecting the specimens to diametral compression in the same apparatus using the 250 Kg beam. The Hounsfield tensometer is classified according to British Standard 1610: 1958 as grade B with a minimum accuracy of 1.5% of the applied load, or 0.3% of the maximum scale reading whichever is the greater. The tensometer load was applied at a crosshead speed of 1 mm/min. by an electric motor. Load values were plotted manually and read to an accuracy of 0.5% of the maximum scale reading.

The maximum value for tensile strength by diametral compression is given as follows:

$$f = \frac{2P}{\pi Dt}$$

where f = the tensile strength (Newtons per mm^2)

P = the load at fracture (Newtons)

D = the diameter of the cylinder (mm)

t = the length of the cylinder (mm)

DISCUSSION - COMPRESSIVE STRENGTH AND TENSILE STRENGTH

It is clear from the compressive strength and tensile strengths that consistent results are difficult to obtain. The mean value for compressive strength at 24 hr. is much lower than those obtained by TAYLOR (1929) but supports his comment "that it is difficult to produce two samples from the same alloy having the same or nearly the same physical properties". The results are in general agreement with those of SCHARFENBERG (1948) and MATHEWSON (1970).

A common procedure when testing brittle materials is to exclude results which fall outwith $\pm 15\%$ of the arithmetic mean of the total number of specimens. In the 24 hr. study the effect of excluding these results outwith $\pm 20\%$ of the original mean reduces the number of specimens from 65 to 30. This reduction altered the original mean value of 120.7 N/mm^2 by a negligible amount.

Compressive strength at 7 days is significantly higher at 158.4 N/mm^2 than the 24 hr. strength.

The exclusion of results outwith $\pm 20\%$ of this mean reduces the number of specimens from 32 to 21, a proportionately smaller reduction than the 24 hr. study. Having examined the analysis of the packing technique on the 24 hr. studies*, each specimen in this study was condensed by 100 ± 5 thrusts of between 35 and 44N (8 and 10 lbf). The coefficient of variation (v) was less than that of the 24 hr. study.

* See Chapter 6 sources of error, p.134

RESULTS

Compressive Strength at 24 hr. - N/mm²

Cylindrical specimens 10 x 5 mm

A. Complete data

Temperature of COPPER AMALGAM

Preparation	n=	\bar{x}	s
150°C	12	113.4	22.1
160°C	8	112.7	50.5
170°C	20	124.2	41.1
180°C	16	118.5	24.1
190°C	9	133.7	34.8
Overall	65	120.7	35.5

v=29.4%

B. Data \pm 20% of above overall mean

150°C	8	116.7	12.2
160°C	NIL		
170°C	9	120.4	14.4
180°C	11	117.9	11.0
190°C	4	-	range 149.8-
			152.3
Overall	30	122.7	15.9

v= 13%

SILVER AMALGAM

C.

New True Dentalloy	6	330	range 309-340
Amalcap	6	354	range 312-378

Compressive strength at 7 days - N/mm^2

Cylindrical specimens 10 x 5 mm

A. Complete data

Temperature of preparation	n=	\bar{x}	s	
160°C	32	158.6	40.3	$v=25.4\%$
% Hg content	32	62.5	2.3	$v=3.6\%$

B. Data $\pm 20\%$ of above mean compressive strength

160°C	21	153.9	26.7	$v=17.4\%$
% Hg content of above specimens		62.8	2.3	$v=3.6\%$

Residual Mercury Content - per cent

	n=	\bar{x}	s	range
24 hr. tensile strength specimens	29	59.24	5.1 ($v=8.6\%$)	53.7-70.4
7 day compressive strength specimens	32	62.5	2.3 ($v=3.6\%$)	59.1-68.0

Tensile Strength (by diametral Compression) at 24 hrs.
- N/mm²

Cylindrical Specimens 10 x 5 mm

COPPER AMALGAM

A. Complete data

Temperature

of preparation

n =

\bar{x}

range

150°C	5	15.3	12-17
160°C	6	19.7	19.23
170°C	6	17.6	16-19
180°C	6	12.7	9-20
190°C	6	18.3	17-19
Overall	29	17.02	4.65

v = 27.3%

% Hg Content

29

59.24

5.1

v = 8.6%

B. Data \pm 20% of above mean tensile strength

	19	17.29	1.7
--	----	-------	-----

v = 9.8%

% Hg content of
above specimens

61.21

5.2

v = 8.4%

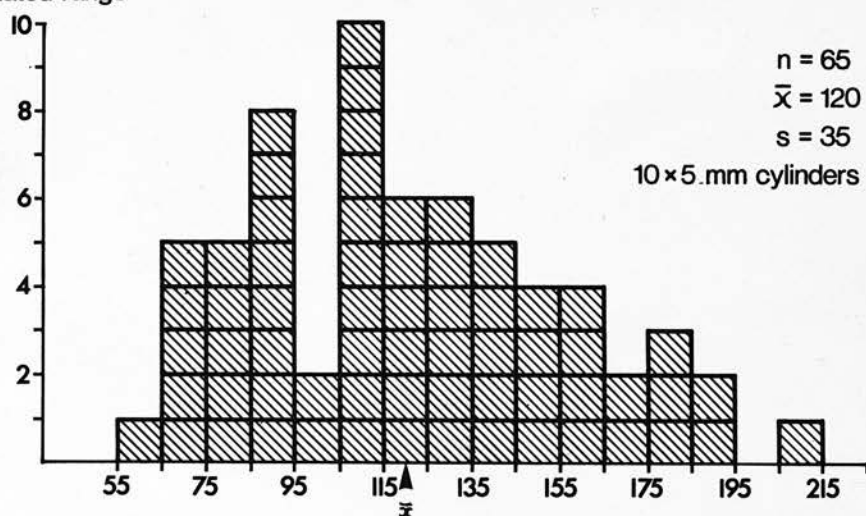
SILVER AMALGAM

C. Amalcap	6	56	range 53 - 59
------------	---	----	---------------

DISTRIBUTION OF COMPRESSIVE STRENGTH OF COPPER AMALGAM

No. of specimens
in stated range

24 HOUR



7 DAY

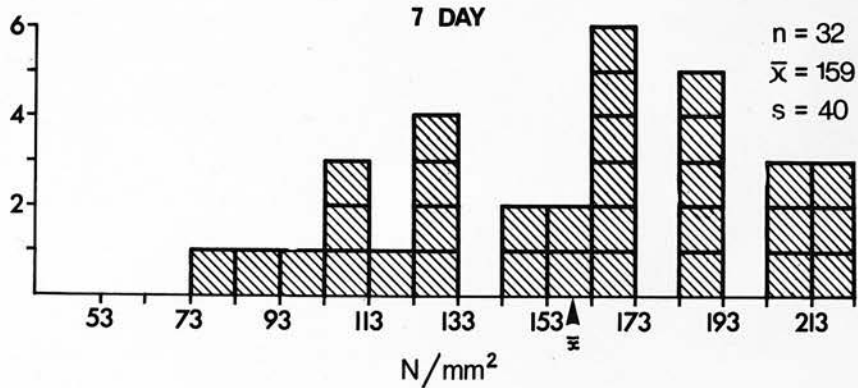


Fig. I.7

91

The distribution of results is shown in Fig. 1.7.

The results of mercury analysis show that there is no relationship between mercury content and the compressive or tensile strengths. However, the coefficient of variation for mercury content for the 24 hr. tensile strength (8.6%) which was completed before the analysis of condensation technique compared with that of the 7 day compressive strength (3.6%) packed by the stricter technique indicate that these strengths may be related to condensation techniques. Both condensation techniques are more exacting than that which could be applied clinically.

VICKERS HARDNESS

It was decided that microhardness in a substance which was not homogeneous and exhibited porosity would be of less importance than a hardness test of a more general and comparable nature. A test which would simulate conditions in the mouth to some extent would be of more practical value than a test which yielded information about the microstructure.

In the Vickers Hardness test an indenter, in the form of a square pyramid with 136° face angles, is pushed into the surface of the prepared specimen at a predetermined load (F) and applied for 10 - 15 seconds. The surface area of the impression is calculated using the arithmetic means (D) of the measurements of the two diagonals. The applied load is divided by the surface area of the impression to give the hardness thus:

$$HV = \frac{2F \sin \theta/2}{D^2}$$

where F = load in kilogrammes-force (Kgf)

d = arithmetic mean of the two diagonals
in millimetres

HV = Vickers Hardness

An "AVERY-6406 Visual Hardness testing machine" was used to carry out the hardness tests. This machine enabled the rate of application of the load (20 kg) to be applied at the same speed for each specimen. The indentation is then projected on a measuring screen which, by means of a micrometer screw enables measurements to be made to an accuracy of 0.001 mm. The screen can rotate to measure the second diagonal.

Cylindrical specimens 10 x 5 mm were used as before. They were polished flat at both ends and three indentations made at each end.

RESULTS

VICKERS HARDNESS

A. Copper amalgam 24 hr.

Temperature of preparation	n=	\bar{x}	range
-------------------------------	----	-----------	-------

160°C	10	96	71-120
-------	----	----	--------

Copper amalgam 1 year

Temperature of preparation	n=	\bar{x}	range
-------------------------------	----	-----------	-------

160°C	10	150	122-183
-------	----	-----	---------

B. Silver amalgam 24 hr.

Room temperature	6	98	93-103
------------------	---	----	--------

DISCUSSION - VICKERS HARDNESS

The Vickers Hardness for silver amalgams and copper amalgams at 24 hrs. would appear to be similar under the conditions of preparation already described, despite a considerable difference in their compressive strengths. The 1 year specimens of copper amalgam were approximately 50% harder than the 24 hr. specimens. Since there is unlikely to be a significant difference in mercury content between the two groups of specimens, it would appear that ageing has taken place which takes considerably longer than 24 hrs. The hardness is not related to the mercury content.

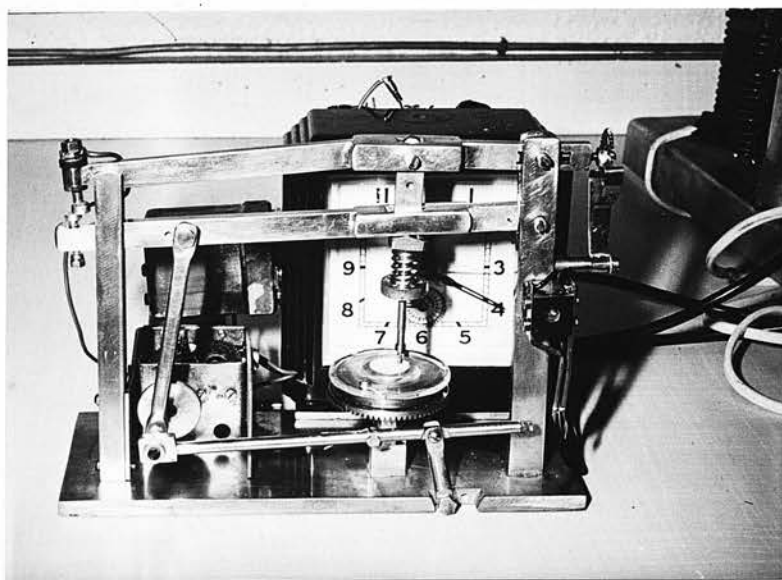


Fig. I-8

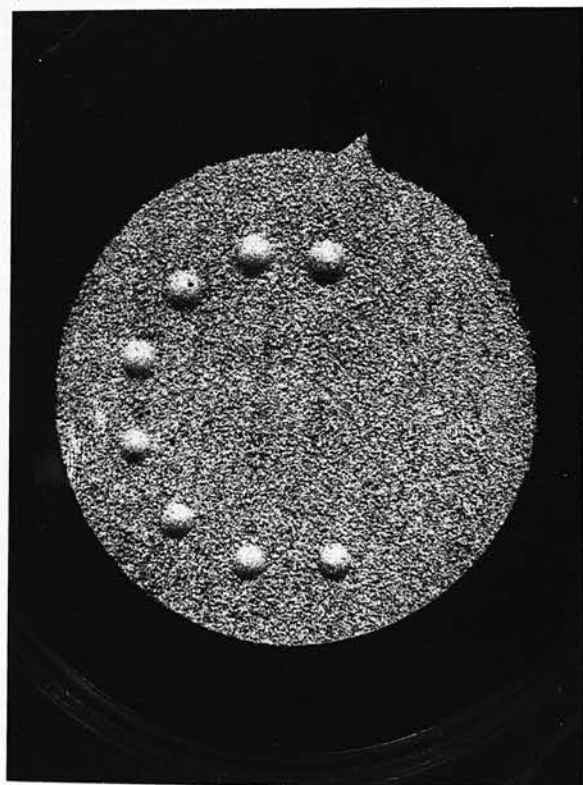


Fig. I-9a

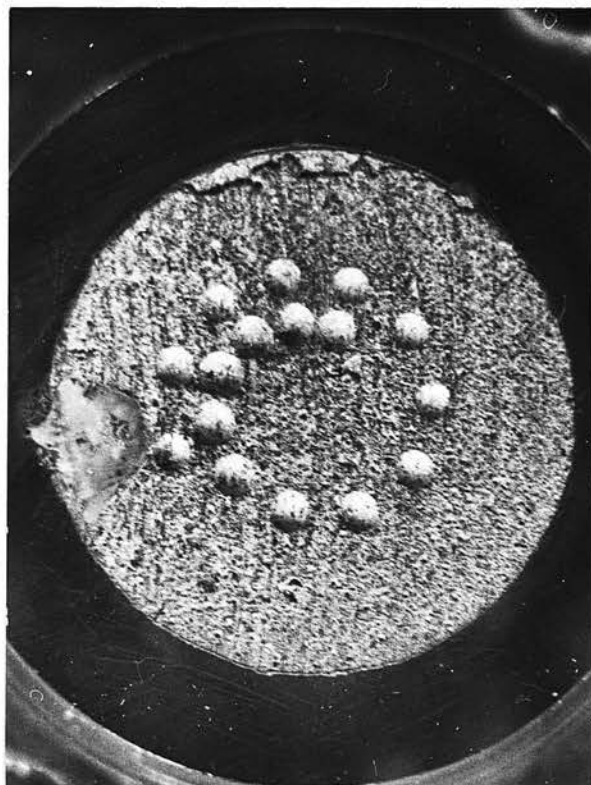


Fig. I-9b

INITIAL SETTING TIME

The aim of this study was to compare the initial setting time of copper amalgam with that of silver amalgam. The initial setting time of amalgam is important. High early strengths are desirable so that the forces of occlusion do not damage the restoration. One of the disadvantages of copper amalgam would appear to be its slow initial setting time.

DEFINITION

In this study the initial setting time is defined as the time beyond which the carving of a silver amalgam ceases to be practical.

APPARATUS

Discs of copper and silver amalgam were made 1.4 mm. in diameter and 2 mm. thick in a special acrylic mould which enabled the surface of the amalgam to be planed flat immediately after packing. The mould containing the freshly packed amalgam was then placed in the apparatus as shown in Fig. 1.8. An indenter ball 1 mm. in diameter and carrying a 1 Kg. load was then applied to the surface of the disc at a constant speed resting on the specimen for 10 seconds. The indenter was removed and the specimen turned automatically to a fresh position and the test repeated. The time between applications could be varied by means of the clock in the apparatus. The clock was started at commencement of trituration.

The apparatus was designed to stop when the indenter ball ceased to penetrate the sample beyond a

predetermined depth. This could be set by means of adjustable contacts which cut off the power supply.

CALIBRATION

A fine grain silver amalgam (New True Dentalloy) which complied with B.S. 2938^{*} was selected as a standard. The clock on the apparatus was started at the commencement of trituration, which was done by hand, and a disc of the silver amalgam produced and installed in the apparatus. The apparatus was then set to indent the specimen continuously each cycle taking 30 seconds while the remainder of the mix was tested for carveability. When carving became difficult, the adjustable contacts were set to stop the apparatus including the electric clock. The indenter made very slight indentation in the silver amalgam disc when carving became difficult (Fig. 1.9a).

The experiment was repeated with a copper amalgam specimen but a 5 minute time lapse was set between each indentation to allow for the extended setting time. The apparatus was also designed to produce a spiral trace to accommodate longer setting times (Fig. 1.9b).

* Specification for Dental Amalgam alloy.

RESULTS

SILVER AMALGAM

Minutes

	n=	\bar{x}	range
New True Dentalloy	6	14	13-16

COPPER AMALGAM

Temperature of preparation - 160°C

Neo Silbrin	7	53 \pm 5	50-70
Cupro Muc	7	58 \pm 5	30-83
Ash's	2	127.5	120 and 135

SUMMARY

It is clear from the above studies and experience gained from handling copper amalgam that the material differs from dental silver amalgam.

The values for compressive strength of copper amalgam at 24 hr. are approximately one third of the values obtained for two dental silver amalgams. At 7 days the value for compressive strength of copper amalgam rises by approximately 50 per cent and the initial setting time was approximately four times as long as the silver amalgam used as a control. These last two properties indicate that the setting reaction is very slow compared with dental silver amalgam. High 1 hour strengths are considered to be important in the development of current dental silver amalgams.

27

The copper amalgams tested in this study all exhibited a setting contraction. The overall average value was 0.13 per cent. An increase in the temperature of preparation of 40°C did not appear to increase the setting contraction.

The fact that silver amalgams exhibit an initial contraction, then expand, as dimensional change on setting is plotted against time, in contrast to the simple exponential curve described by copper amalgams, again demonstrates a fundamental difference between the two materials.

It was not found possible to control all the variables in the production of the test cylinders of copper amalgam but results seemed to indicate that the compressive strength and the tensile strength of the material, which were extremely variable, were not related to the residual mercury content of the test cylinders.

In addition, metallurgical examination of the copper amalgam specimens showed that the distribution of copper in any given sample is uneven and this characteristic together with that of random porosity could account for the inconsistent results of some of the mechanical tests.

From the mechanical tests described the average values for the following properties were obtained:-

Dimensional change on setting	=	0.123% contraction
Compressive strength - 24 hr	=	120 N/mm ²
Compressive strength - 7 day	=	159 N/mm ²
Tensile strength - 24 hr.	=	17 N/mm ²
Vickers Hardness - 24 hr	=	96

CHAPTER 2

ADAPTATION STUDY

ADAPTABILITY AND MICROLEAKAGE STUDIES OF COPPER AMALGAM AND SILVER AMALGAMS

ADAPTATION

It was decided to compare the adaptation of copper amalgam with some silver amalgams currently available.

METHOD and MATERIALS

The method devised by McHUGH (1955) was estimated to provide a test of adaptation which would simulate conditions likely to be encountered when packing a retention groove cut by a $\frac{1}{2}$ round bur. The dies are easily manufactured and are entirely suitable for subsequent techniques. They are transparent acrylic blocks with cylindrical cavities 8 mm deep and 4 mm in diameter. The cavities have a standard metric thread (No. 9, 0.75 mm pitch) cut into the walls (Fig. 2.1).

After the amalgams were packed and examined in the dies they were removed and subjected to a quantitative examination by means of the Scanning Electron Microscope (SEM), (BOYD, 1969).

The following amalgams were used in this study:

Brand	Type
A. New True Dentalloy	Fine grain
B. Amalcap Capsule Alloy	Preamalgamated
C. Luna Atomic non-zinc	Spherical particle
D. Kerr's Spheraloy	Spherical particle
E. Solila Fine Grain	Fine Grain
F. Cupro-muc	Ready made copper amalgam

Four specimens of each amalgam were prepared in accordance with the manufacturers' instructions, making a total of 24. Alloys "A, C and E" were triturated by hand and alloys "B" and "D" were triturated in the "Silamat" high energy amalgamator for 5 and 8 seconds respectively. 7 - 10 increments of 0.1 gm. were required to fill each test cavity to excess with silver amalgam. In "A" the first two increments contained unexpressed mercury as recommended by the manufacturer. No mercury was expressed in "B, C and D".

The copper amalgam was prepared as described in Chapter 1. In this experiment 5 pellets were heated to 160°C and triturated by hand. Each specimen of copper amalgam required 12 - 14 increments to fill the test cavity, almost double the number required for the silver amalgam specimens.

Packing was carried out by hand and by Bergendal vibrator driven at 2,000 revolutions per minute, 2 specimens by each technique. A 2 mm. diameter cylindrical condensing tip was used throughout the experiment for both hand and vibrator packing so that the spherical particle alloys could be more easily handled.

The hand packed specimens were produced using apparatus which recorded the total number of thrusts between 35 and 44N required to produce each specimen. This was not possible using the vibrator. Each sample was overpacked and the excess removed.

After 48 hours the specimens were sectioned

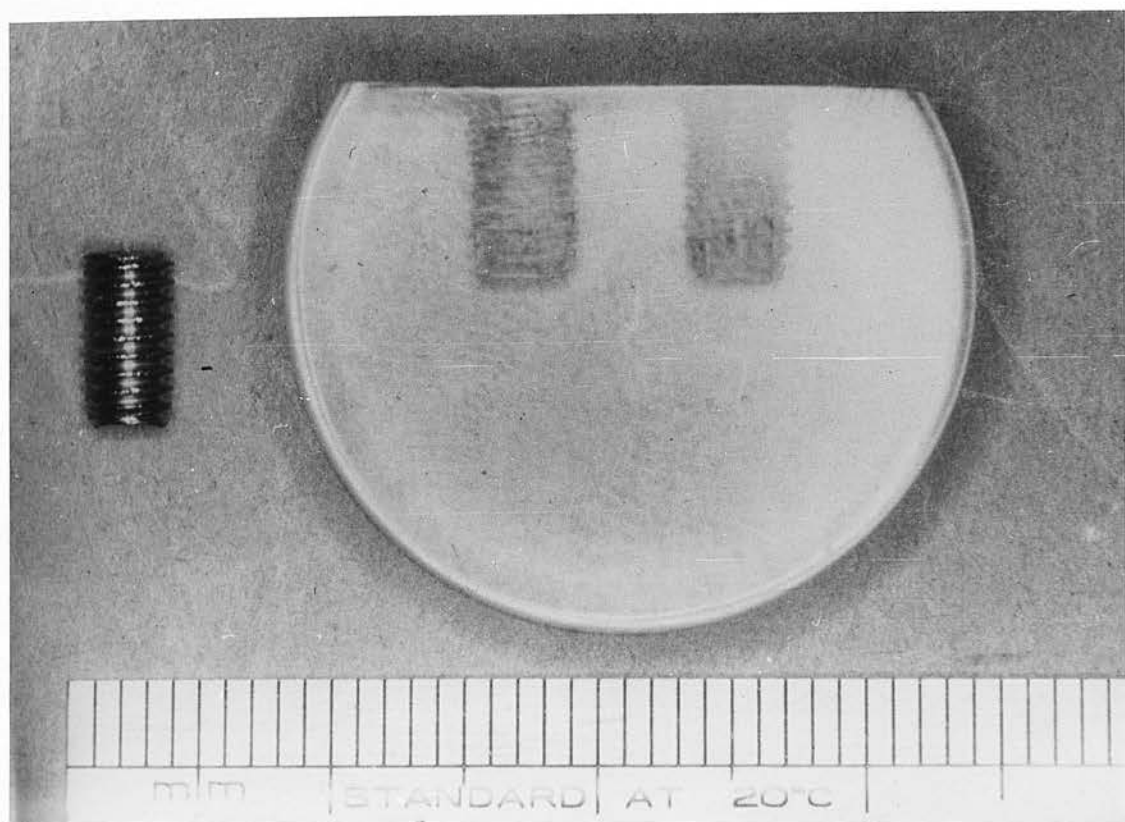
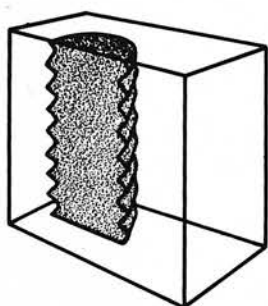


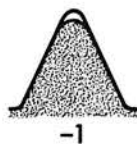
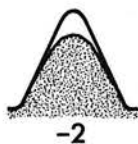
Fig. 2·I

Acrylic mould and resultant pattern for examination by scanning electron microscope, after McHugh (1955).

ADAPTATION SCORE

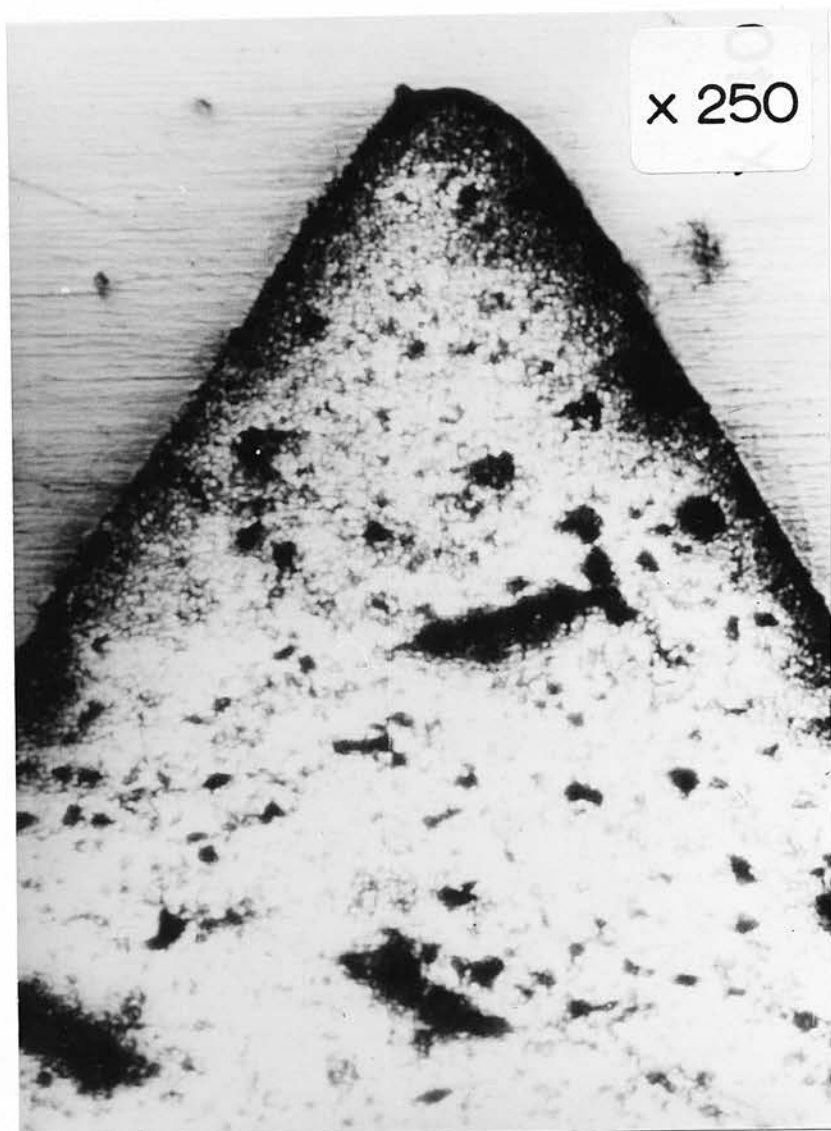


sectioned acrylic block
containing
polished amalgam sample



$$\frac{\text{TOTAL FAULTS}}{\text{NO. OF THREAD PROFILES} \times 5} = \text{deficiency score}$$

Fig. 2•2



optical
section

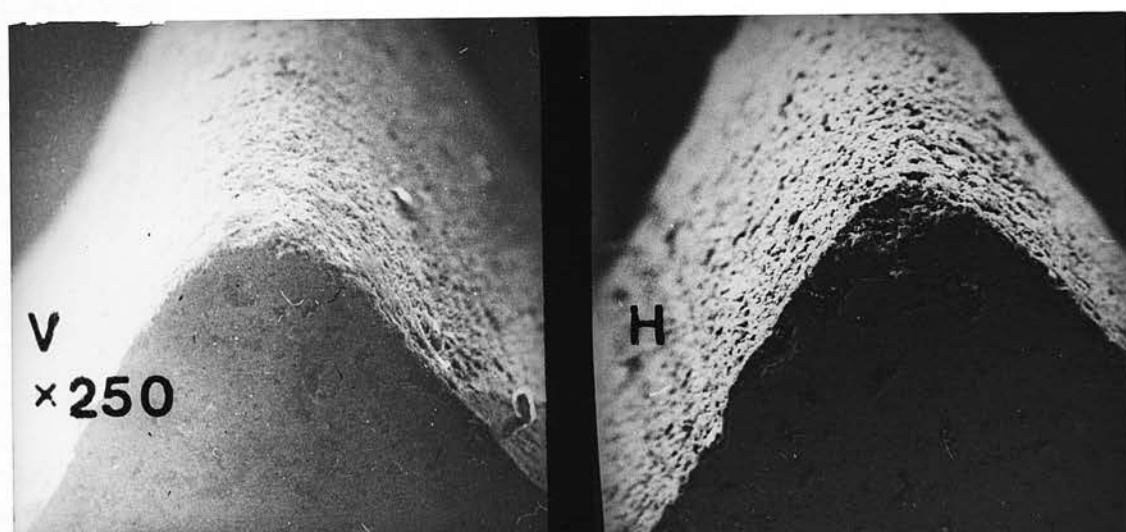
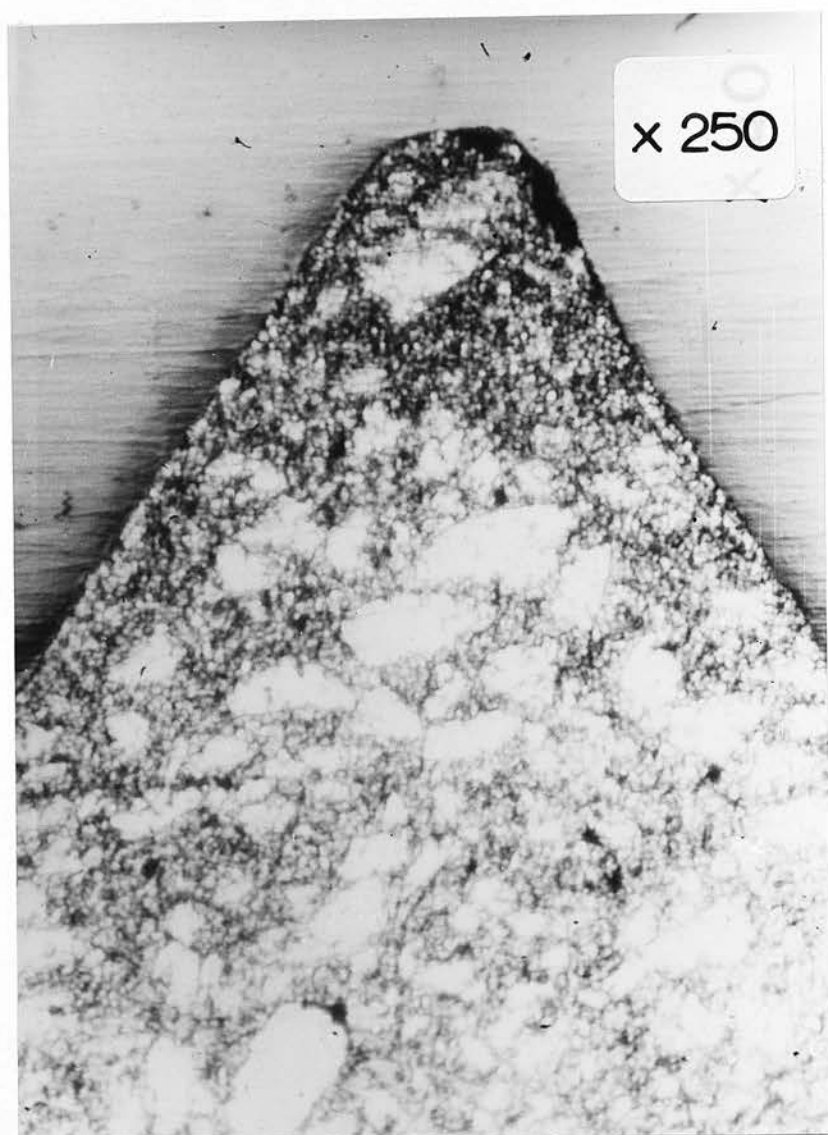


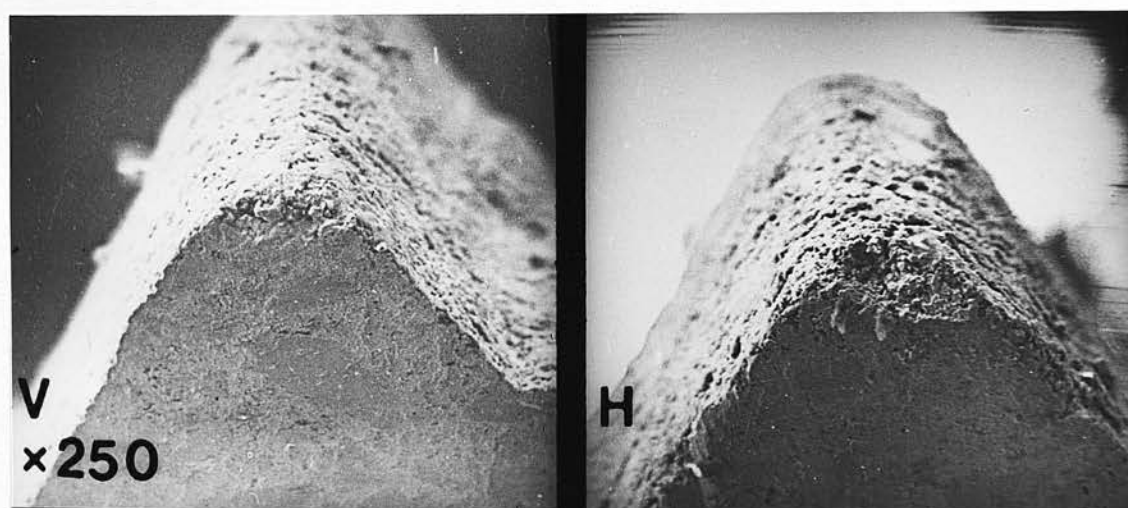
Fig. 2.3

SEM profiles

Copper amalgam (cupro-muc).



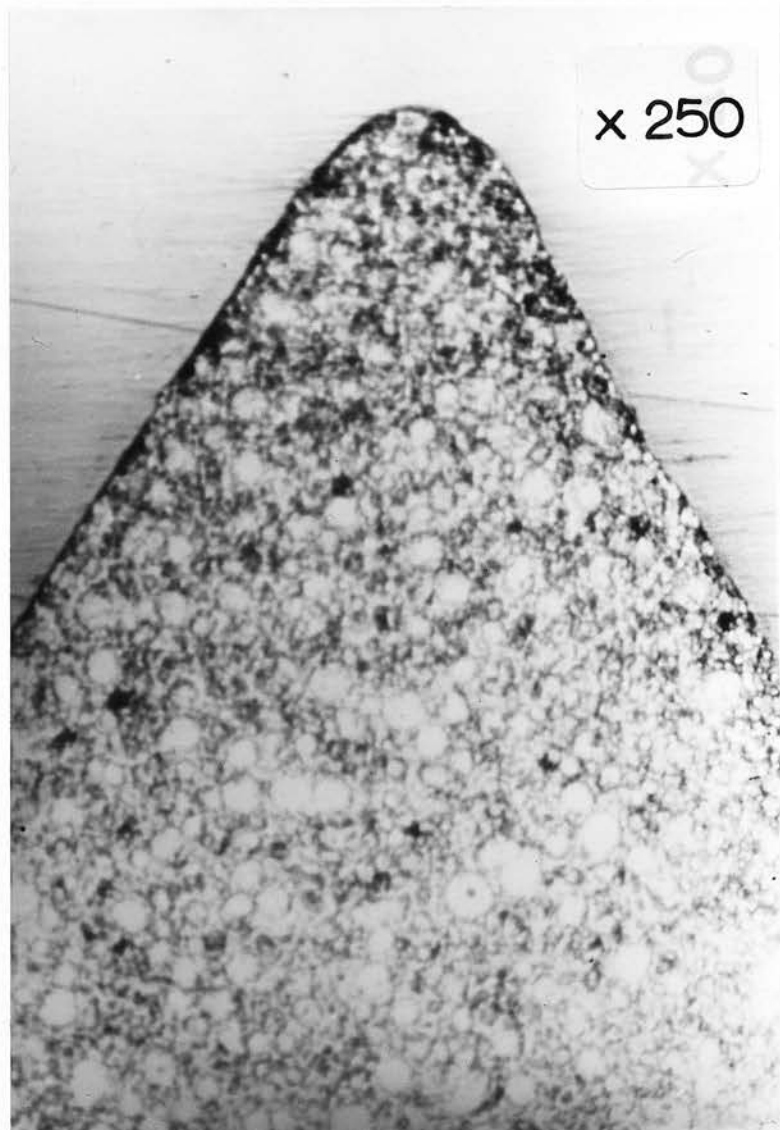
optical
section



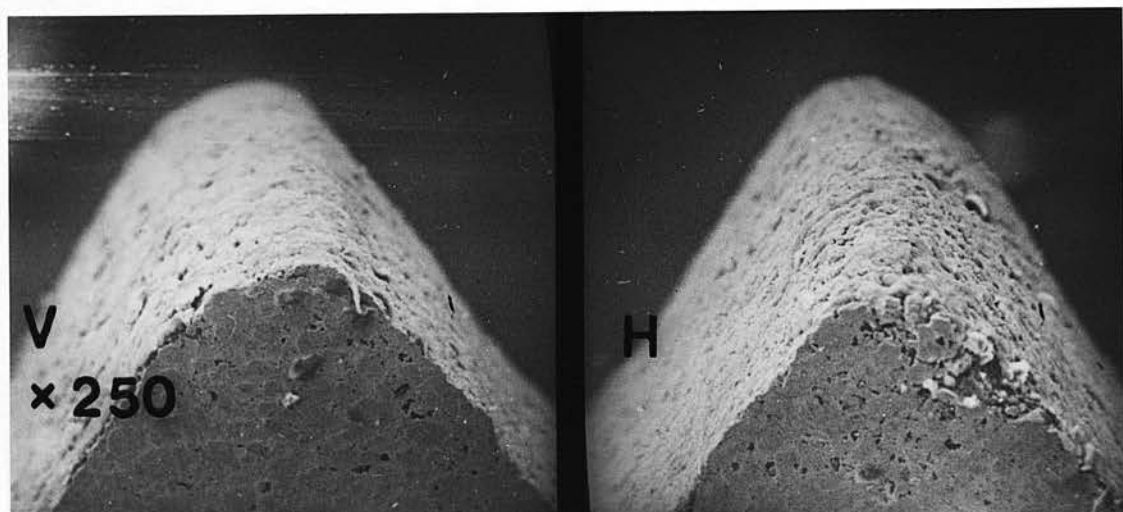
SEM profiles

Fig. 2.4

'Amalcap' - preamalgamated.

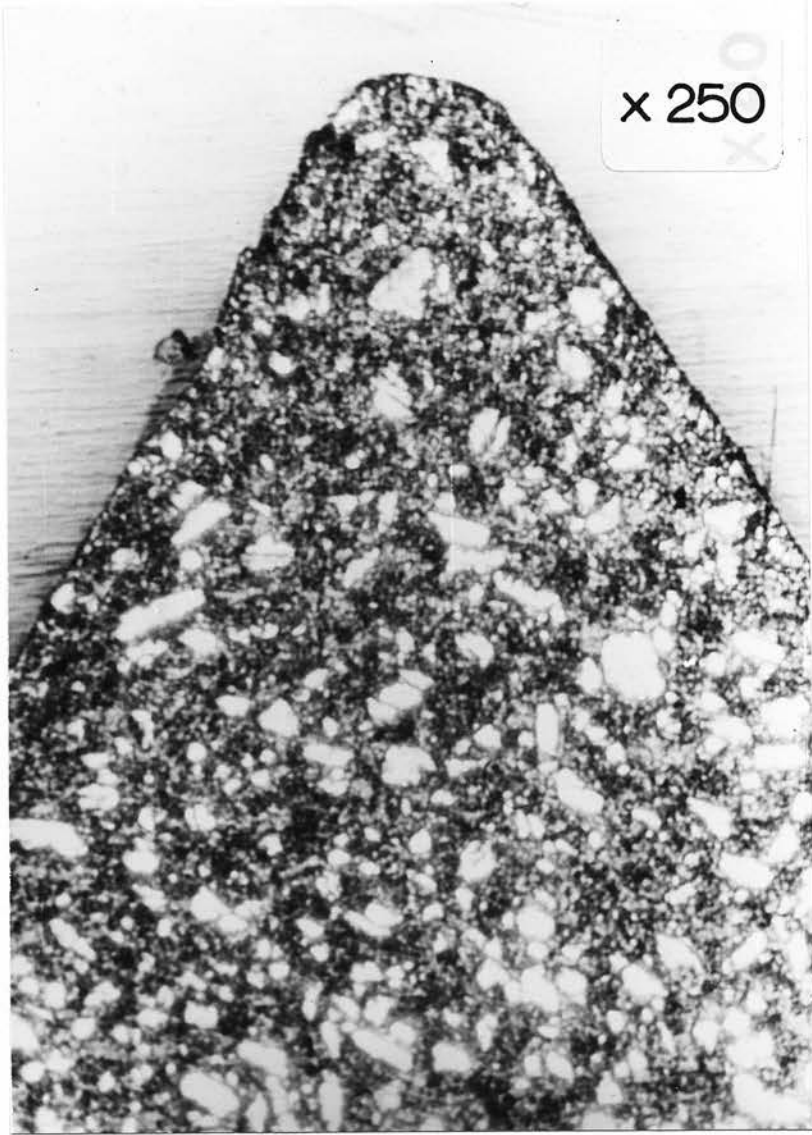


optical
section

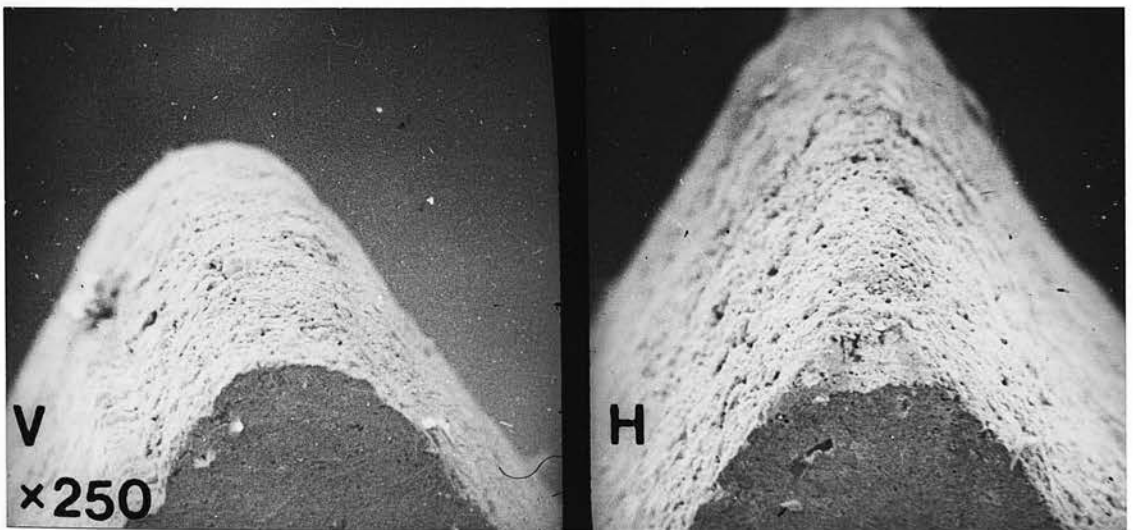


SEM profiles

Fig. 2.5
Kerr's Spheralloy.



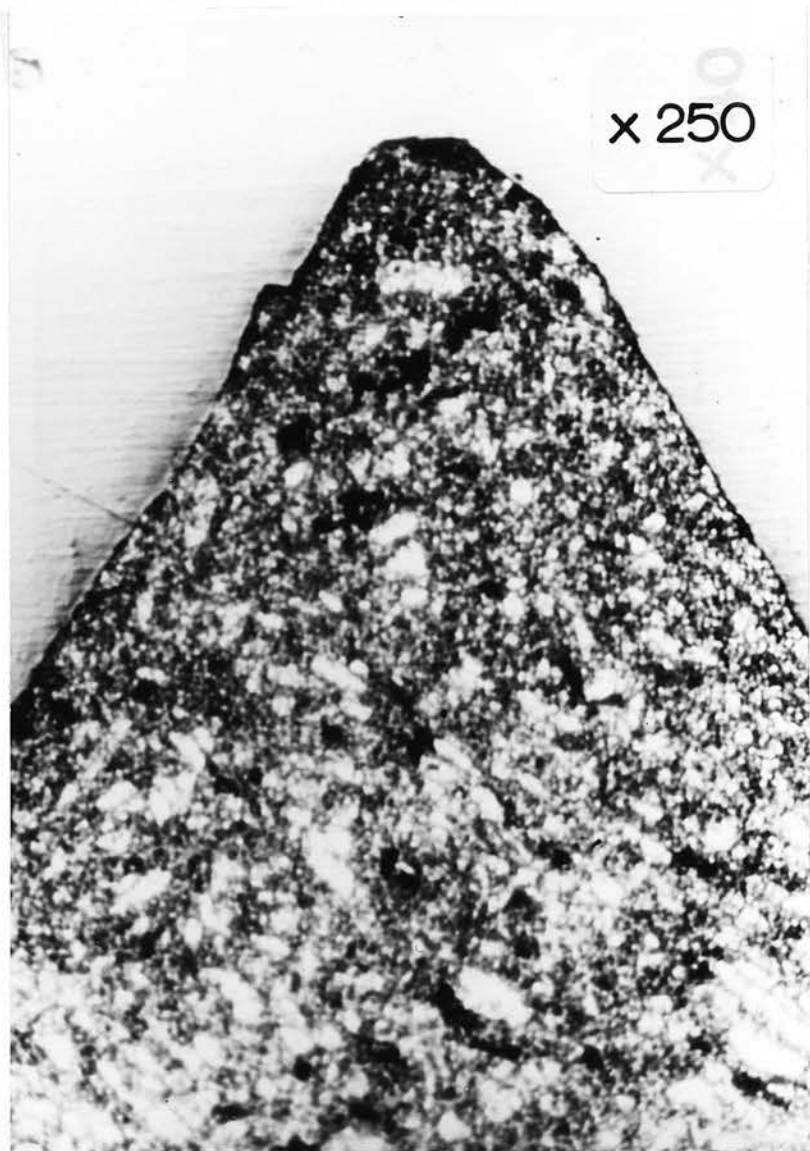
optical
section



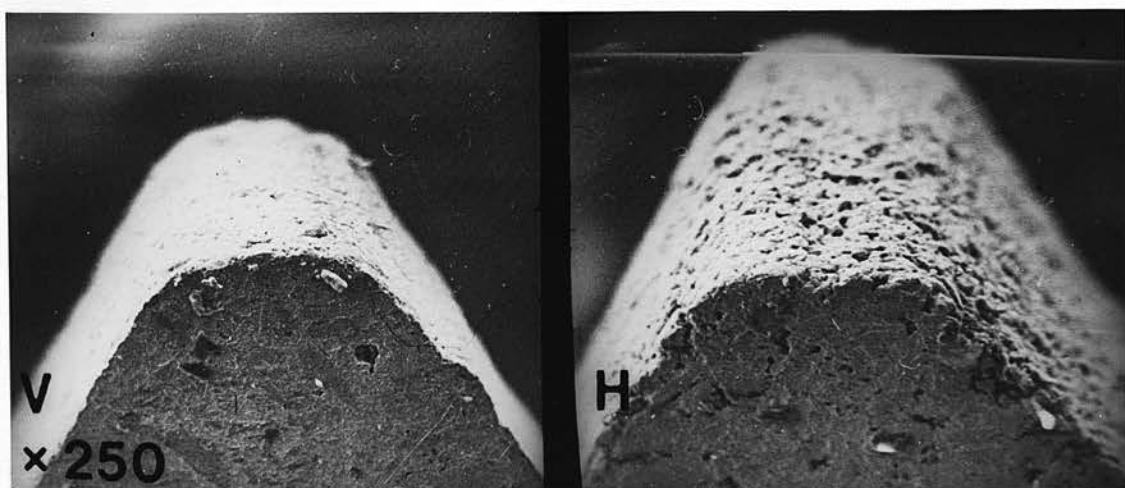
SEM profiles

Fig. 2.6

Solila - fine grain.



optical
section



SEM profiles

Fig. 2.7

New true Dentalloy

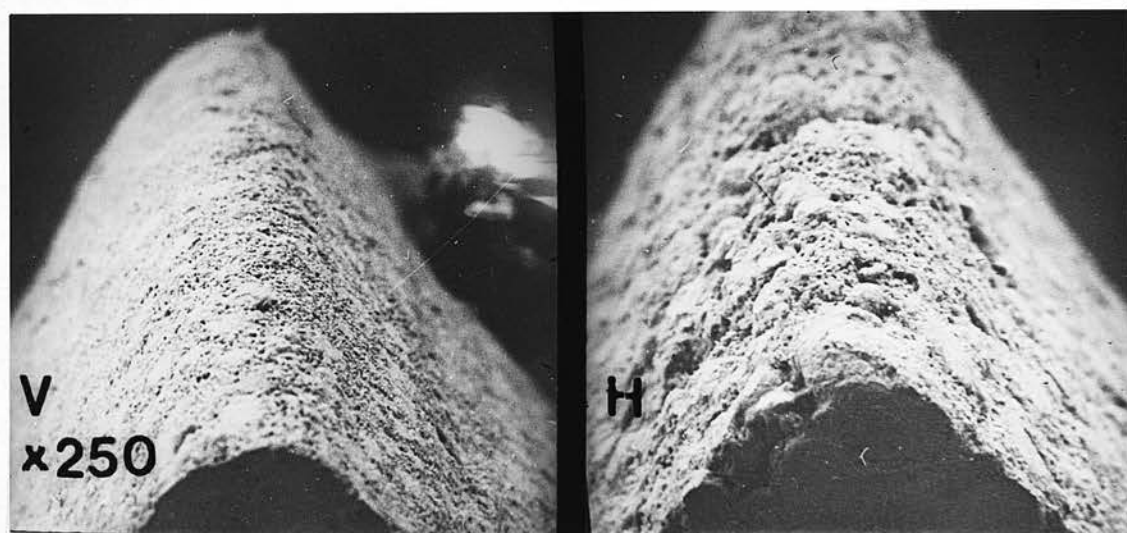
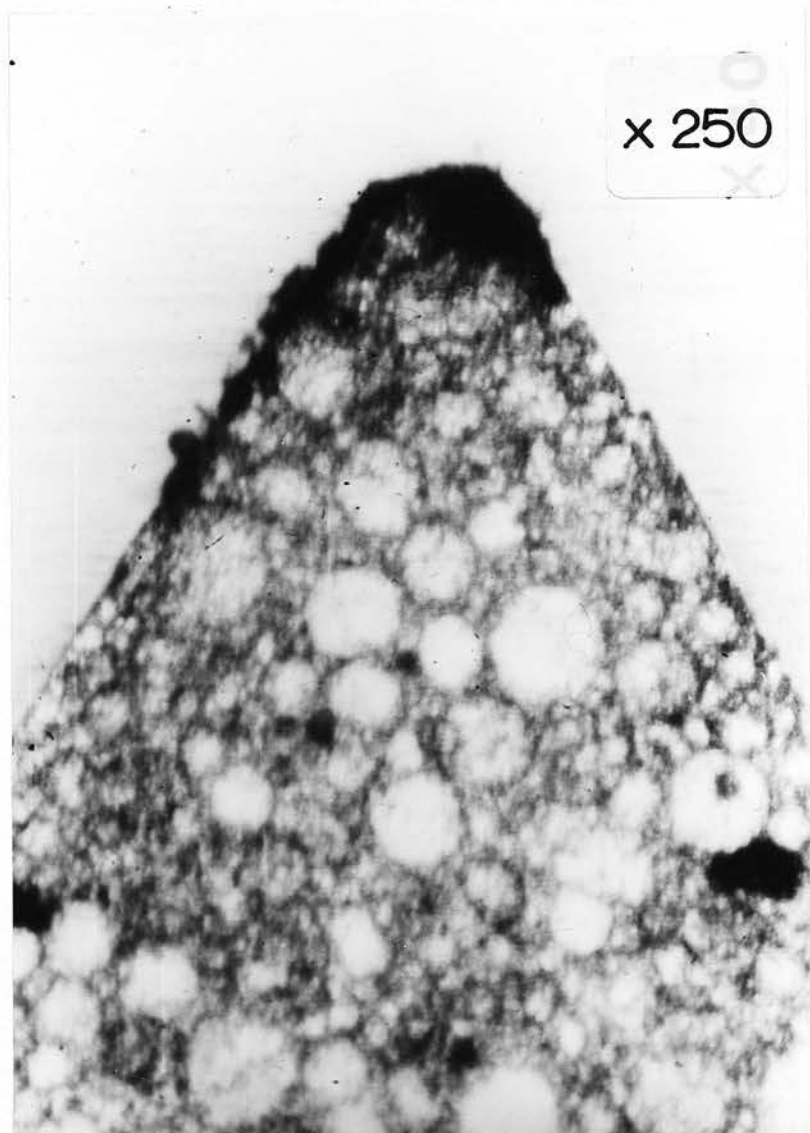


Fig. 2-8

SEM profiles

Luna atomic - non zinc

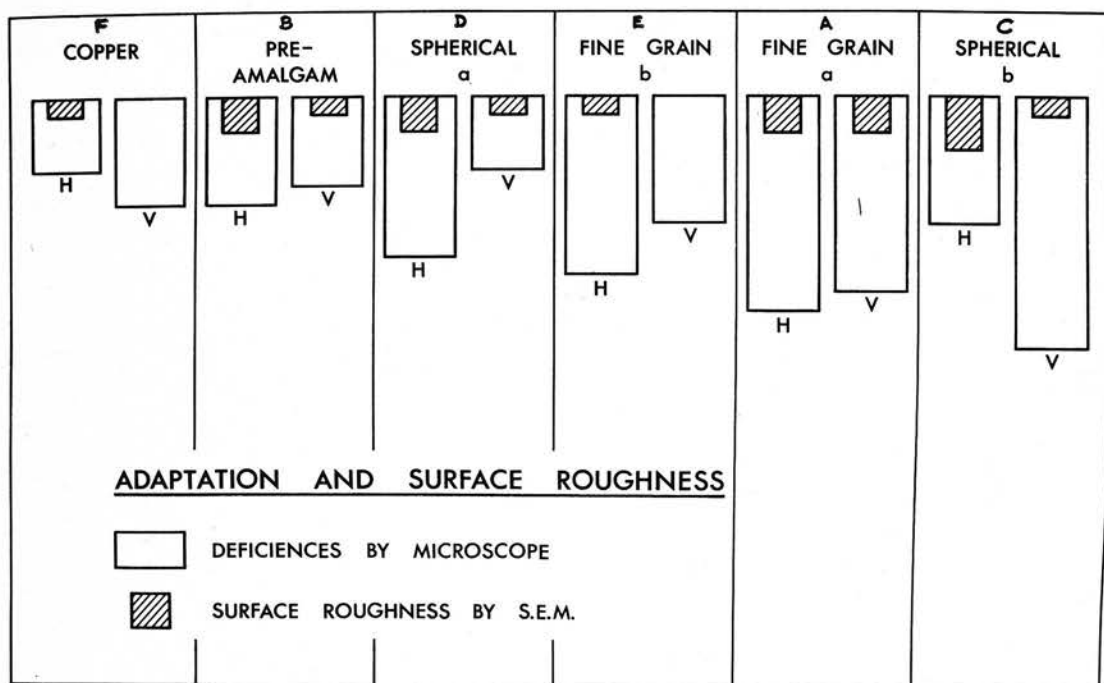


Fig. 2.9.

The comparative adaptability of I copper amalgam and 5 currently used silver amalgams.

using a fine metal saw cooled under cold tap water. The specimens were then prepared as for metallurgical examination using diamond pastes. The silver amalgams were etched using an 8 per cent solution of potassium cyanide containing 1 gm. of iodine per 25 cm.³ applied by swabbing, followed by immersion in a 50: 50 solution of 10 per cent Ammonium hydroxide and 10% Ammonium ferri-cyanide (WING, 1965). The effects of oxidation were sufficient to act as an etchant for the copper amalgam specimens.

Each separate thread recess on the section was examined by optical microscope and assessed according to Fig. 2.2. The total number of faults was then related to the number of recesses, which varied from 7 to 9 per side of the section depending on the depth to which the tap had reached on each mould. This figure was then expressed as a percentage so that the results were comparable. The results are shown in Fig. 2.9.

The specimens were then carefully removed from the acrylic moulds and mounted edgewise so that the quality of adaptation on the thread profile could be examined by scanning electron microscope (SEM). Thus only one side of each of the 2 specimens was available for examination. The specimens were cemented to the SEM studs using cellulose acetate adhesive and electrical conductivity was ensured by using metallic paint over the adhesive touching the stud and the specimen. The

resultant photographs, which require no special interpretation, were simply arranged in order of adaptation by brands. Adaptation differences within brands were graded according to the surface roughness of the profile as shown in the SEM photographs (Fig. 2.3 - 2.8).

The results are presented as an inverted histogram (Fig. 2.9) which represents relative deficiencies in adaptation. The "H" refers to hand packed specimens and the "V" to specimens packed by the Bergendal Vibrator using small increments.

DISCUSSION

Results for copper amalgam and the amalgam made from a preamalgamated silver alloy are in agreement with those of JØRGENSEN (19⁶5) and show that copper amalgam exhibits the closest adaptation of those amalgams examined. They also show that the adaptation of silver amalgams is related to the original alloy particle size, with one exception, the preamalgamated fine grain alloy. This supports the statement made by WING (1966) that the amount of each alloy particle reacting with the mercury is small as judged from a study of the particle sizes and shapes in the set amalgam compared with those of the original alloy.

Copper amalgam differs from silver amalgam in that it does not exist as a particle/matrix system but simply a mass of finely divided copper suspended in mercury which combines slowly to form a bimetallic compound.

The properties of adaptation are, therefore, different from those of silver amalgam and would appear to depend on the degree of plasticity of the amalgam and condensation technique. The plasticity of copper amalgam prepared for use in the mouth is ultimately proportional to the mercury content.

It is clear from these amalgam studies that adaptation is a more important factor in microleakage than dimensional change on setting. It could even be argued on the basis of particle size alone, that an amalgam with poor adaptability which exhibited a setting expansion would be more susceptible to microleakage than a well adapted amalgam which showed an equivalent setting contraction.

MICROLEAKAGE

A small number of microleakage tests on copper amalgam and silver amalgam restorations in extracted teeth were carried out to investigate the relationship to their adaptation properties described above.

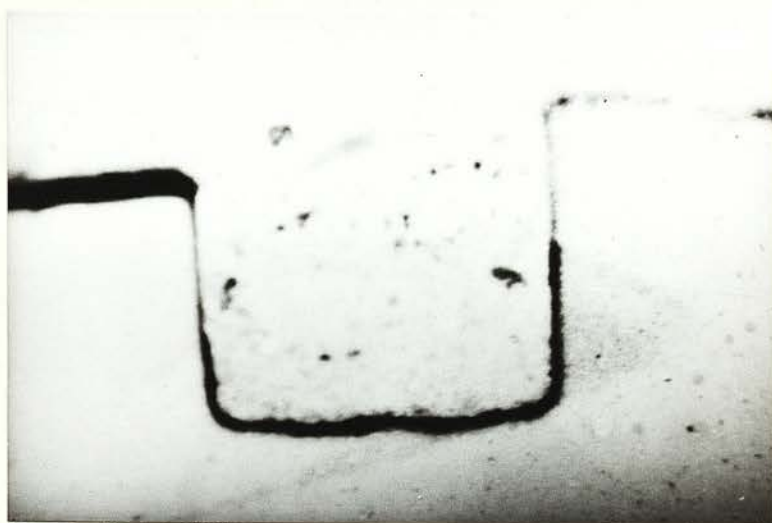
METHOD and MATERIALS

Single surface cavities (Class V) were prepared in 20 freshly extracted teeth. The cavity margins were finished using a flat finishing bur (ALLAN, 1968). Restorations of copper amalgam and silver amalgam (10 each) were placed without linings. The fine grain silver amalgam (A) "New True Dentalloy" was selected from its position relative to copper amalgam on the histogram, (i.e., as showing comparatively poor adaptation, Fig. 2.9).



penetration of toluidene blue

(a) Photomicrograph x30



penetration of 535

(b) Autoradiograph.

The teeth were then painted with cellulose acetate varnish to within 1 mm of the cavity margin to prevent ingress of the tracer solution from any other source.

The teeth were then placed in a 10% solution of aqueous sodium sulphide, containing S^{35} as sodium sulphate (50 microcuries per cm^3) and 3.8% toluidene blue, (LYELL *et al.*, 1964). The teeth were stored at $37^{\circ}C$ for two weeks.

The teeth were then removed from the solution and washed thoroughly in tap water. They were dried in air then embedded in clear acrylic resin*. The teeth were then sectioned using a water cooled diamond saw so that the plane of section divided the tooth longitudinally through the restoration. The sections were polished as for metallurgical examination. The sections were photographed and corresponding autoradiographs made by simply clamping small intra-oral film against the embedded section. The exposure time in this study was between 6 and 8 weeks.

RESULTS AND DISCUSSION

All the restorations showed signs of microleakage. It was not possible to detect any difference in marginal leakage between copper amalgam restorations and silver amalgam restorations. Information gained from the use of the toluidene blue dye was slightly better than that of the autoradiographic technique in so far as the dye showed a greater degree of penetration into the dentine. (Fig. 2.10a and b).

* "Marco" resin 5B 28c

104

The results are in agreement with those of GOING et al., 1960b, who also showed that microleakage was present in both types of amalgams tested. Clearly the use of a "caulking" agent seems fundamental to the prevention of fluid exchange at the margins of amalgam restorations.

CONCLUSIONS

Copper amalgam adapts to the cavity wall better than the silver amalgams used for comparison in this study. The fact that copper amalgam differs in structure chemically and physically from dental silver amalgam is offered as an explanation for this finding.

The closeness of adaptation of both amalgams was insufficient to prevent microleakage.

CHAPTER 3

ANIMAL STUDY

A COMPARISON OF THE EFFECTS OF COPPER AMALGAM AND SILVER AMALGAM ON THE RAT MOLAR PULP

Introduction

SHROFF (1952) has stressed the importance of histological studies of the pulpal responses to filling materials as well as a study of the physical and chemical properties. FARRIS and GRIFFITH (1949) state that the histology and physiology of rat molar teeth is similar to human teeth. HOFFMAN and SCHOUR (1940) studied the development of the rat molar in detail and used these teeth to study pulpal response to various stimuli. MAURICE and SCHOUR (1954), KAKESHASHI (1965 and 1969), ROWE (1967) and BERMAN and MASSLER (1968) also used the rat molar in studies of pulpal response to various filling materials.

The purpose of this present study was to investigate the histological changes in the rat molar pulp following the insertion of a copper amalgam restoration, and compare these changes with those following the insertion of silver amalgam. The pulp reactions sometimes associated with cavity preparation are indistinguishable from those associated with filling materials and so every effort was made to reduce trauma during cavity preparation.

Preliminary trials showed that occlusal cavities were unsuitable as intercuspal cavities give a false impression of depth and ROWE (1967) has reported a high proportion of lost amalgam restorations in occlusal cavities in rat molar teeth caused by a weakening of the tooth structure after cavity preparation.

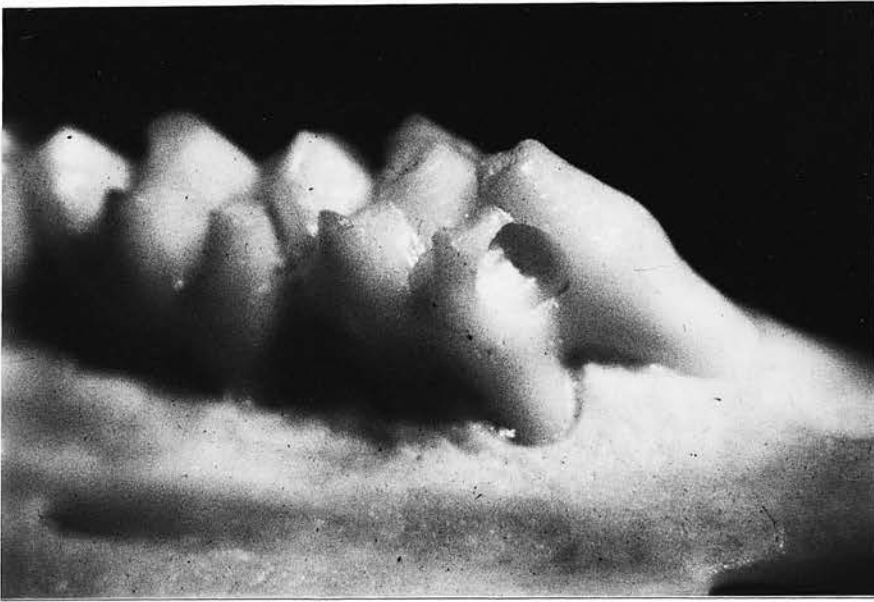


Fig.3-1.

Position of cavity on mesio-lingual surface of rat molar tooth (after Paynter and Wood, 1955)

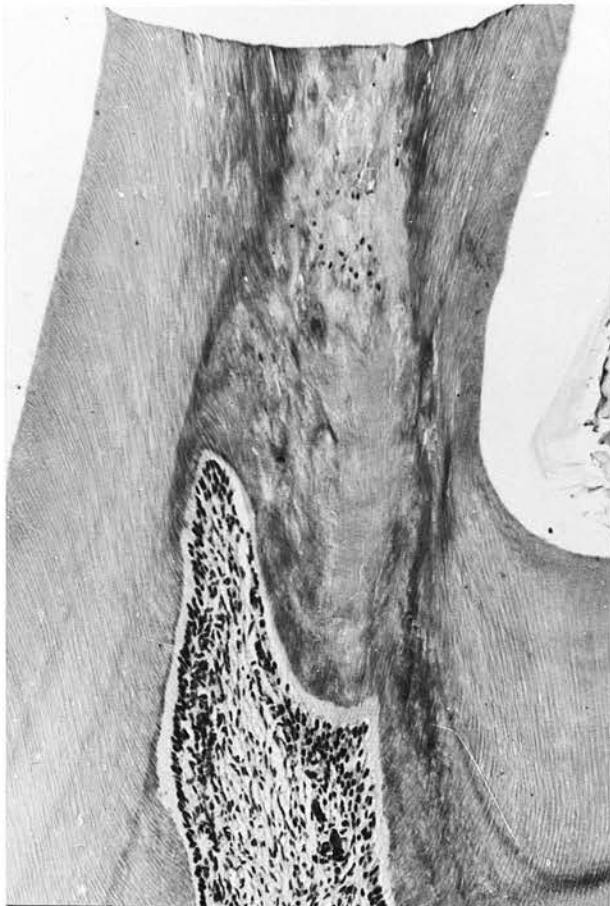


Fig.3-2.

Physiological secondary dentine in normal (i.e. not subjected to any operative procedure) rat molar.

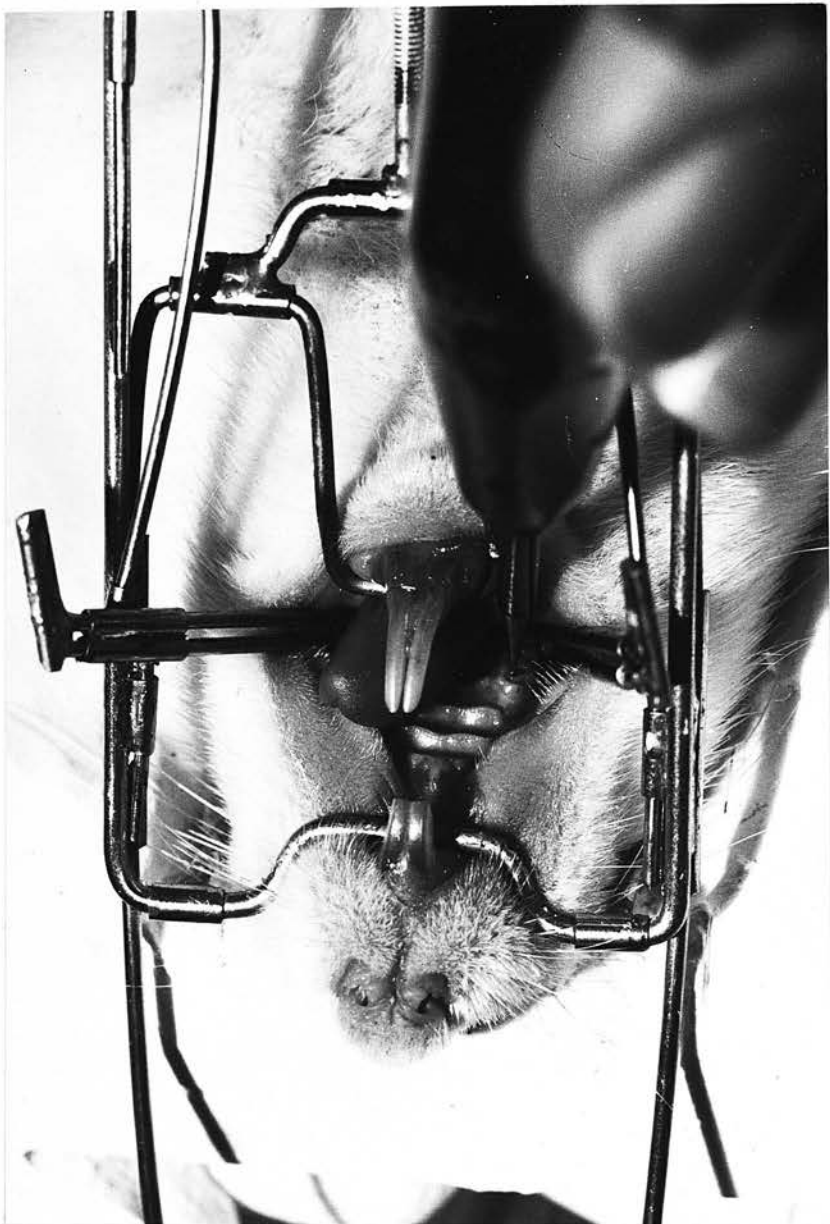


Fig.3-3.

View of operating position showing specially designed adjustable gag with cheek retractors.



Fig.34.

Small 6 volt electric motor adapted
to fit conventional straight dental
handpiece.

It is important that a sufficient number of teeth with retained restorations are available for evaluation of pulpal response. The cavities in the present study were, therefore, placed on the mesiolingual aspect of the maxillary first molar tooth (Fig.3.1) a site also used by PAYNTER and WOOD (1955). In this location any localised dentine reaction could be distinguished from the two types of secondary dentine normally found in the rat molar, namely pulp horn secondary dentine which forms under the cusps at 35 - 45 days of age and pulpal roof or floor secondary dentine which starts to form at 135 days of age (Fig.3.2). HOFFMAN and SCHOUR (1940).

Silver amalgam was used for comparison under similar conditions.

MATERIALS AND METHODS

1. The Experimental Material

87 female, random bred, albino rats, 120 days old were used for this study. In these animals attrition and the formation of pulp horn secondary dentine was found to be constant. Anaesthesia was administered by intraperitoneal injection of nembutal sodium (3 gm per 100 gm body weight). Each animal was then placed on its back in a plaster of paris holder (Fig.3.3). The mouth was held open by a special gag with cheek retractors which was attached to the holder. A dental chip syringe was used to blow away debris and used to control saliva. Cavities were prepared in the first maxillary molar teeth of each animal, one was filled with copper amalgam, the other with silver amalgam.

The rats were fed the normal axoid modified diet 41-b with water ad lib. They were sacrificed after periods of 1 month, 2 months, 3 months and 4 months and the restorations examined in situ by stereomicroscope to record loss or retention of the restoration.

2. Cavity Preparation

The cavities were cut by hand to a depth of approximately 0.25 mm using half the diameter of the 005 I.S.O. round bur as a guide. A straight dental handpiece was used, driven by a high quality 6 volt d.c. electric motor at 1,000 revolutions per minute (Fig.34). The cavity was then undercut with an inverted cone bur, 0.520 mm in diameter. An increase in depth was avoided as far as was possible during this procedure.

3. Preparation of the Amalgam

The copper amalgam was heated to a temperature of 160°C in the apparatus previously described and triturated by hand. The silver amalgam was mixed in a "Wig-L-Bug" automatic amalgamator for 10 seconds. Excess mercury was removed in both cases. The amalgams dispensed from a specially made carrier, were condensed using a probe tip flattened to a diameter of 0.45 mm. One silver amalgam and one copper amalgam restoration were inserted in unlined molar cavities on opposite sides of the maxilla. To simplify laboratory procedures each type of amalgam restoration was allotted to the same side of the maxilla in a given group of animals.

4. Histological Technique

After sacrifice the rats' heads were removed and fixed in 4% buffered formaldehyde. During fixation the

jaws were propped open by cotton wool swabs soaked in fixative. After 7 days the mandibles tongues and posterior portions of the skulls were trimmed off and discarded. After a further 3 days' fixation the maxillary tissues were decalcified in 10% formic acid in 8% sodium citrate. The end point of decalcification was determined by the radiographic method of BRAIN (1966). At this stage the maxillae were divided sagittally and excess tissue removed leaving the molar segment intact. After embedding in paraffin wax serial sections of each specimen were cut at 5 micrometres parallel to the median plane incorporating the three molar teeth. (No attempt was made to relate the plane of section to the axes of the cavities.) To avoid damage to the microtome knife, restorations still retained within the paraffin wax were removed when they became visible in the block during sectioning. The sections were stained with haematoxylin and eosin and Van Gieson's stain method for collagen.

RESULTS

Of the 175 amalgam restorations placed in the molar teeth of rats 22.8% of the copper amalgams and 50.6% of the silver amalgams were not retained. This was chiefly due to excessive attrition. The results for each experiment are summarised overleaf in TABLE 1.

A SUMMARY OF PULP HISTOLOGY UNDER RETAINED RESTORATIONS

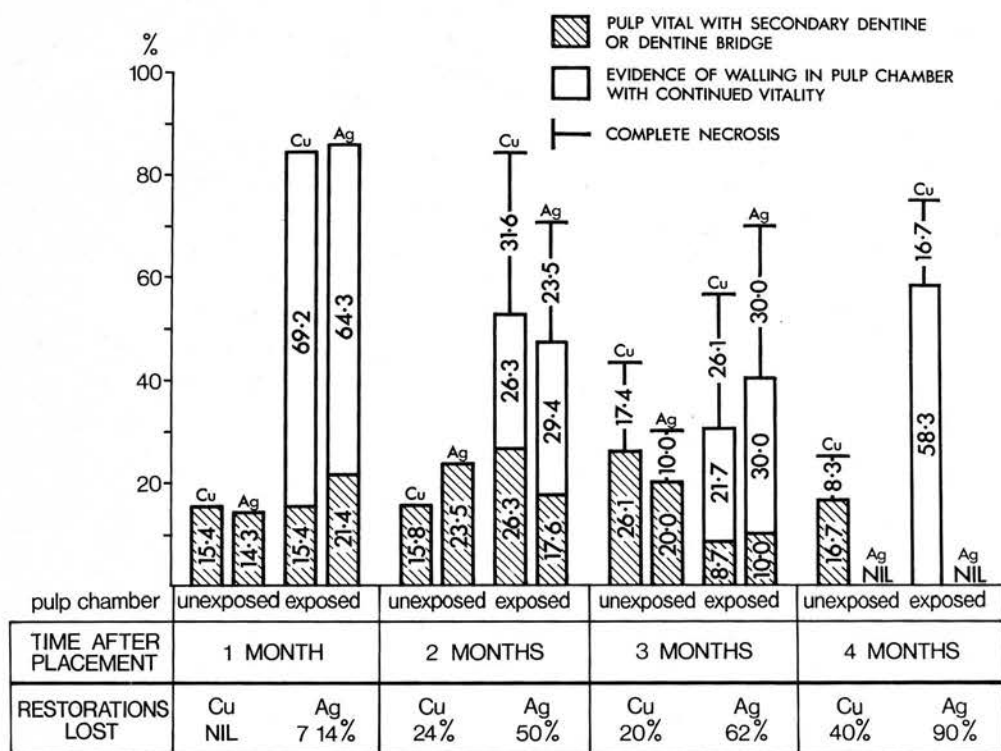
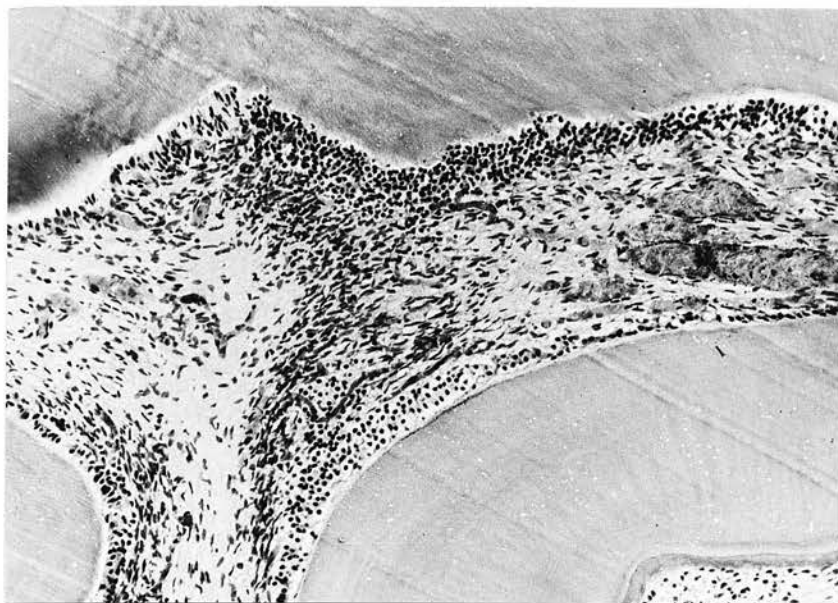
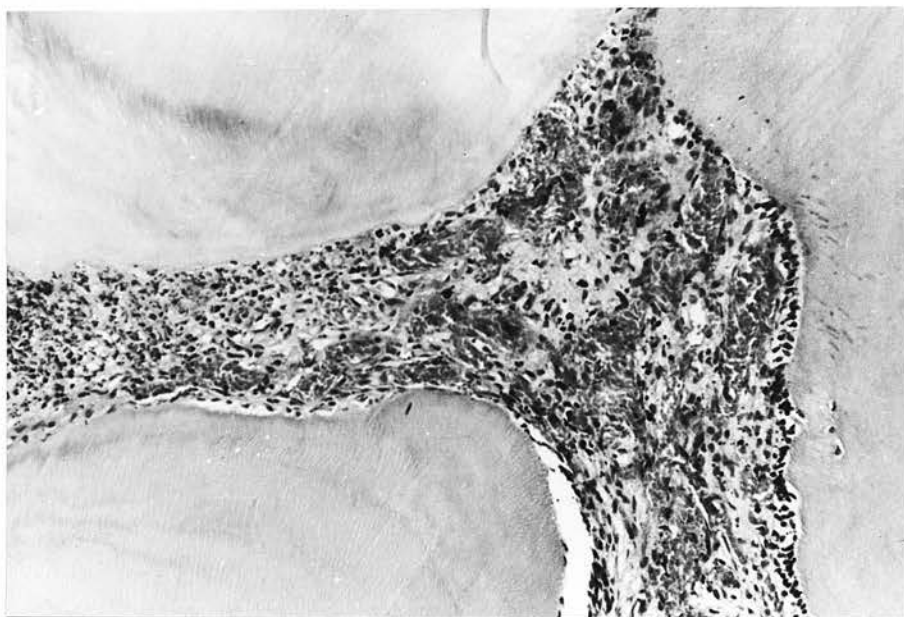


Table I.



H. & E. x120.

Fig. 5. Normal rat pulp. Oblique cutting of odontoblasts, gives at first sight, simulates an inflammatory cell infiltration.



H. & E. x150.

Fig. 6. Pulp with silver amalgam one month. The pulp horn to the left was exposed and the photograph shows marked hyperaemia and inflammation (both estimated at + +) in the adjacent pulp horn.

TABLE 1

NUMBER OF RESTORATIONS LOST

No. of rats in each sample n	Time after placement of restoration	Copper amalgams LOST	Silver amalgams LOST
14	1 month	NIL	1 (7.14%)
*24 (copper 25)	2 months	5 (24%)	7 (50%)
29	3 months	6 (20%)	18 (62%)
20	4 months	9 (40%)	18 (90%)

*one silver amalgam was omitted

RETAINED RESTORATIONS - THE HISTOLOGICAL EFFECTS ON THE DENTAL PULP

The following results are based on examination of those teeth in which restorations were retained for the duration of each experiment.

The term "dentine bridge" is used to describe the formation of dentine within the pulp horn associated with the cavity. The term "walling" is used to describe the formation of dentine elsewhere in the pulp chamber which separates the remaining vital pulp tissue from an area of pulp necrosis related to the exposure. A complete summary is given in Table 2 and a detailed categorisation is given in subsequent tables.

The inflammatory reaction in the pulp was estimated by the degree of hyperaemia and inflammatory cell infiltration, both features being estimated from visual examination of the slides and an impression recorded as -ve, +ve, ++ve, etc. (Fig.3-5 and 6).

COPPER

	1/12	2/12	3/12	4/12
Hyperaemia	- 8 + 7	-11 + 2	-16	- 9 + 1
Inflammation	-11 + 4	-11 + 1 ++1	-15 ++1	- 9 + 1

SILVER

Hyperaemia	-10 + 2 ++2	-11	- 6 + 1	
Inflammation	-10 + 2 ++2	-11	- 7 + 1	

TABLE 3 - UNEXPOSED PULPS

Unlined copper amalgam and silver amalgam restorations placed in the first maxillary molar teeth of the rat where the CAVITY is cut in DENTINE without pulp exposure.

The number of pulps in the above category which showed evidence of continued VITALITY with the formation of SECONDARY DENTINE at the end of each experiment.

	Copper	%	Silver	%
1 month	2/13	15.38	2/14	14.28
2 months	3/19	15.78	4/17	23.5
3 months	6/23	26.08	2/10	20.00
4 months	2/12	16.66	NIL	-
	$\bar{x} =$	18.47	$\bar{x} =$	14.44

TABLE 4 - EXPOSED PULPS

Unlined copper amalgam and silver amalgam restoration in the first maxillary molar teeth of the rat where the pulp was SURGICALLY EXPOSED at the time of cavity preparation.

- A. The number of pulps in the above category which showed evidence of DENTINE BRIDGING associated with the cavity and remaining VITAL pulp tissue at the end of each experiment.

	Copper	%	Silver	%
	n		n	
1 month	2/13	15.38	3/14	21.42
2 months	5/19	26.31	3/17	17.64
3 months	2/23	8.69	1/10	10.00
4 months	NIL	-	NIL	-
	$\bar{x} =$	12.5	$\bar{x} =$	12.26

- B. The number of pulps in the above category which showed evidence of WALLING not directly associated with the cavity and remaining VITAL pulp tissue at the end of each experiment.

	Copper	%	Silver	%
1 month	9/13	69.23	9/14	64.28
2 months	5/19	26.31	5/17	29.41
3 months	5/23	21.73	3/10	30.00
4 months	7/12	58.33	NIL	
	$\bar{x} =$	43.9	$\bar{x} =$	30.9

TABLE 4a - EXPOSED PULPS

A + B

The number of pulps categorised in TABLE 4 which showed evidence of DENTINE BRIDGING associated with the cavity or WALLING not directly associated with the cavity with remaining VITAL pulp tissue at the end of each experiment.

	Copper	%	Silver	%
1 month	11/13	84.61	12/14	85.71
2 months	10/19	52.63	8/17	47.05
3 months	7/23	30.43	4/10	40.00
4 months	7/12	58.33	NIL	-
	$\bar{x} = 56.5$		$\bar{x} = 43.19$	

TABLE 5 - EXPOSED PULPS

Unlined copper amalgam and silver amalgam restorations in the first maxillary molar teeth of the rat where the pulp was surgically exposed at the time of preparation.

The number of pulps in the above category which showed COMPLETE NECROSIS OF THE PULP TISSUE at the end of each experiment.

	Copper	%	Silver	%
1 month	NIL/13	NIL	NIL/14	NIL
2 months	6/19	31.57	4/17	23.52
3 months	6/23	26.08	3/10	30.00
4 months	2/12	16.66	-	
	$\bar{x} = 18.57$		$\bar{x} = 13.38$	

TABLE 6 - UNEXPOSED PULPS

REMAINDER

Dental pulp of the 1st maxillary molar teeth of the rat which showed COMPLETE NECROSIS when the cavity was cut into dentine without surgical exposure and the restoration retained for the duration of the experiment.

	Copper	%	Silver	%
1 month	NIL		NIL	
2 months			1/17	5.88
3 months	4/23	17.39	1/10	10.00
4 months	1/12	8.33	-	

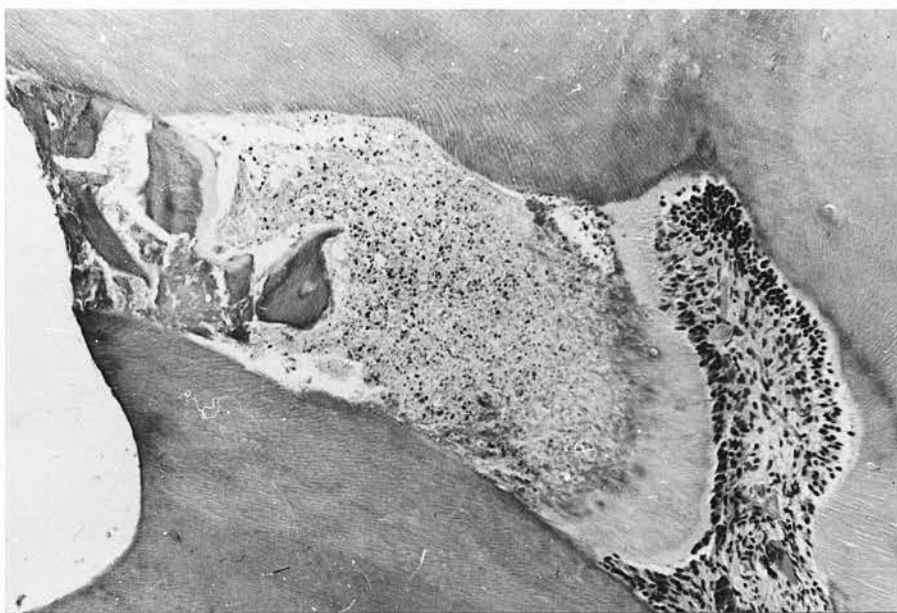


Fig. 7. Low and medium power photographs of a Rat Molar with exposed pulp, the cavity filled with copper amalgam and examined after one month. Both photographs show an area of tissue necrosis beneath the exposure, a dentine bridge, and a relatively normal, vital pulp.

H. & E. x60 and x120.

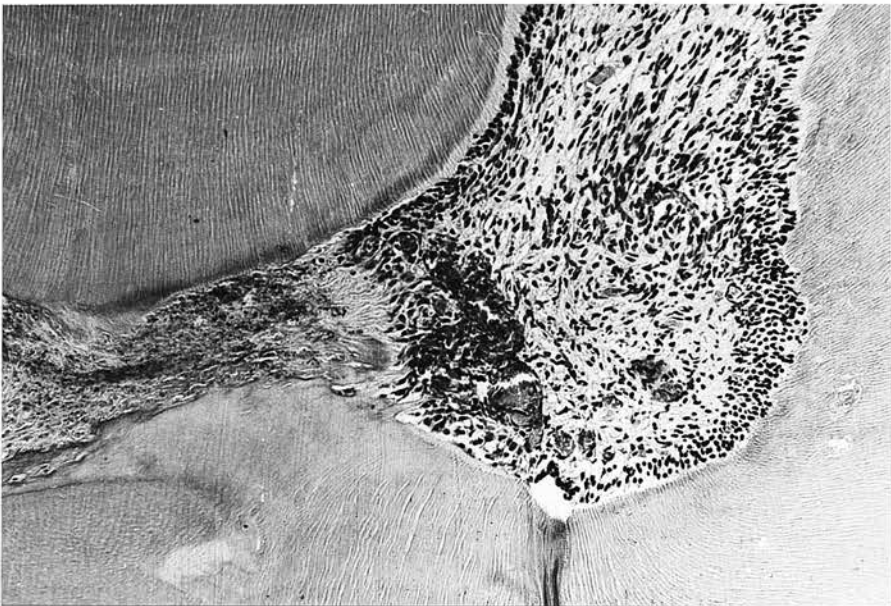
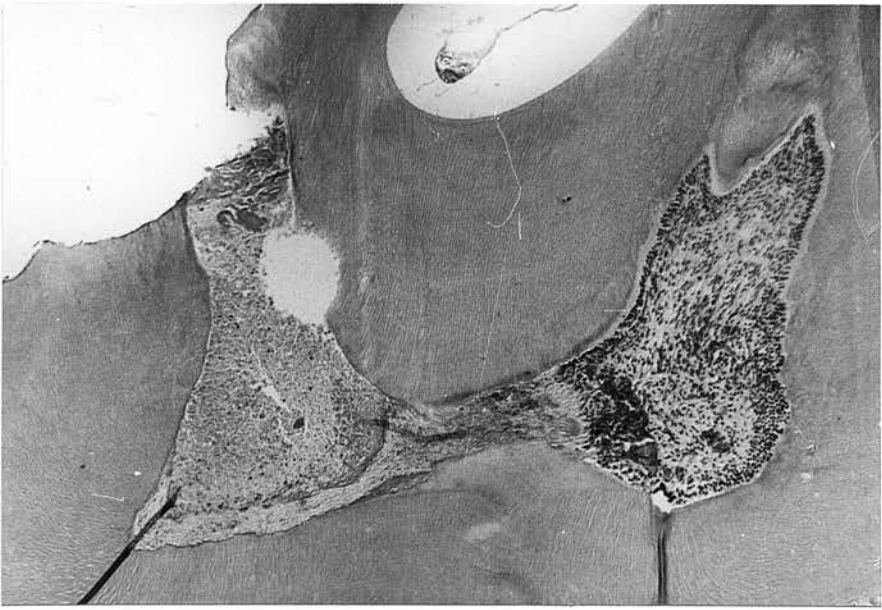
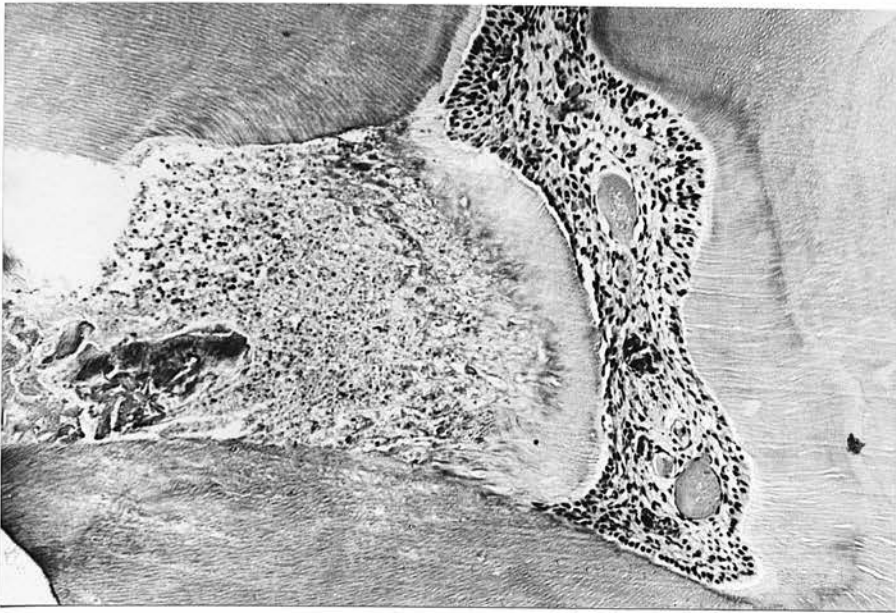


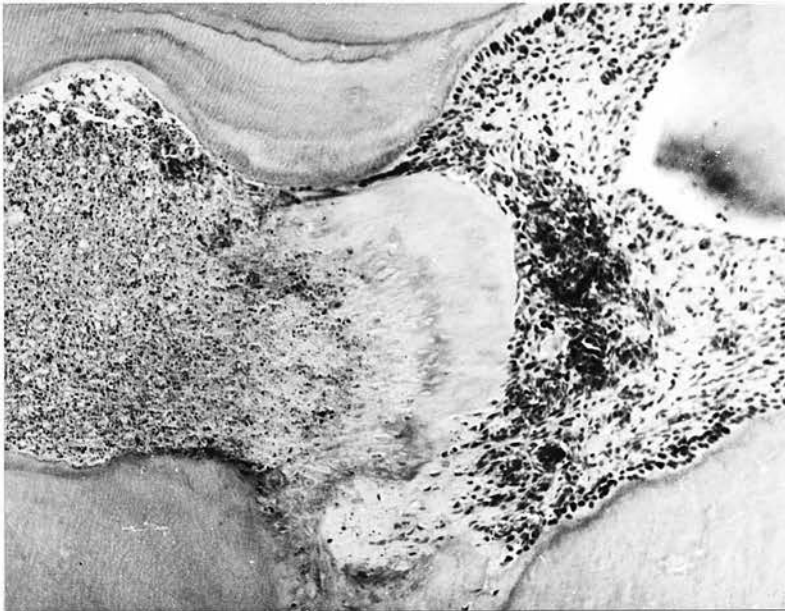
Fig. '8. Low and medium power photographs of a Rat Molar with exposed pulp, the cavity filled with copper amalgam and examined after one month. Both photographs show an area of necrosis extending from the exposure across to the other pulp horn, where a thin wall of dentine has formed. The vital pulp tissue adjacent to the wall of dentine shows slight hyperaemia and inflammatory cell infiltration.

H. & E. x60 and x120.



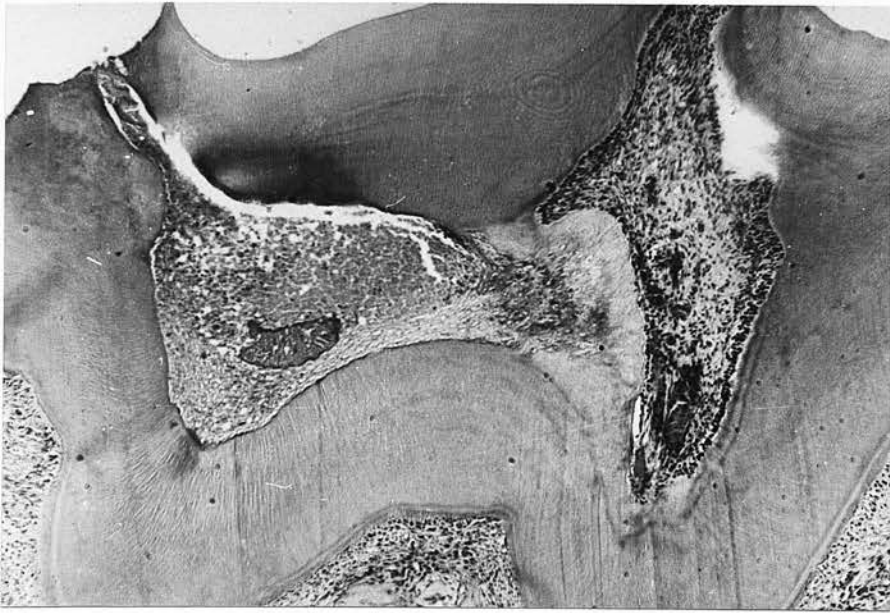
H. & E. x 120

Fig. 9. Rat Molar with exposed pulp, the cavity filled with copper amalgam and examined after one month. The photograph shows an area of tissue necrosis beneath the exposure with a well formed dentine bridge at the base of the exposed pulp horn.

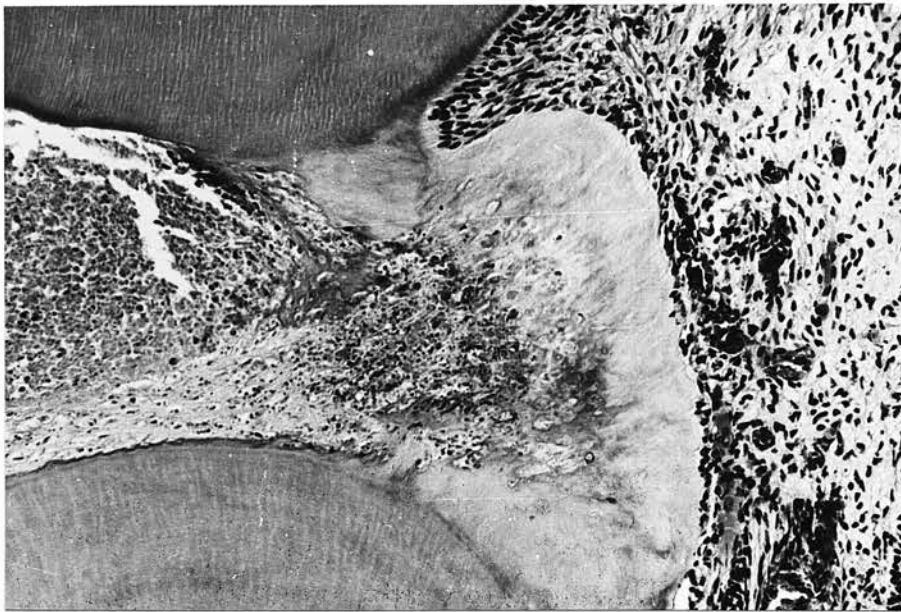


H. & E. x 120.

Fig. 10. Rat Molar with exposed pulp, the cavity filled with copper amalgam and examined after one month. Tissue necrosis beneath the exposure has extended across to the other pulp horn, where a well formed wall of dentine has formed around the necrotic tissue. The vital pulp tissue shows marked inflammatory cell infiltration adjacent to the dentine wall.

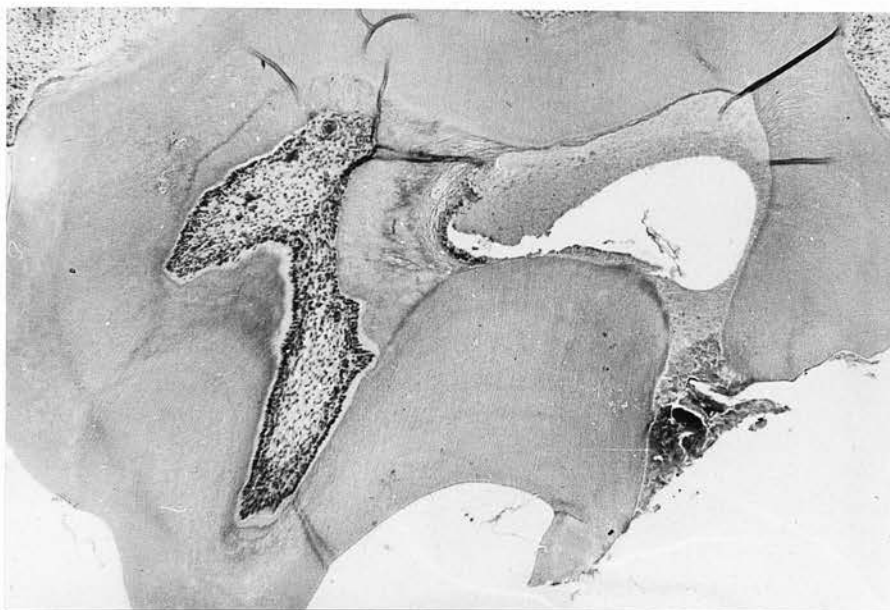


H. & E. x60.



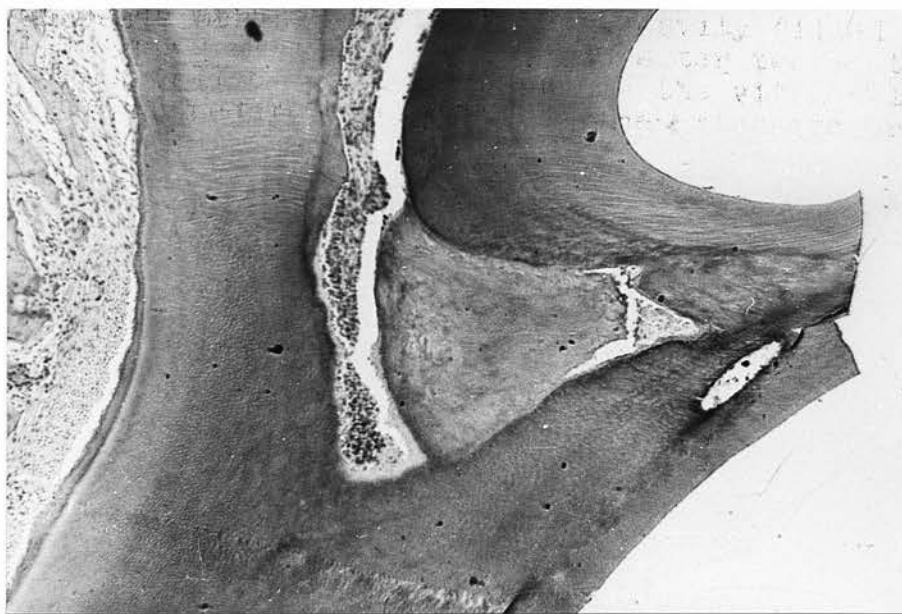
H. & E. x 150.

Fig. II. Low and medium power photographs of a Rat Molar with exposed pulp, the cavity filled with copper amalgam and examined after one month. Both photographs show a well formed wall of dentine in the adjacent pulp horn, separating the slightly inflamed pulp from the extensive area of necrosis beneath the exposure.



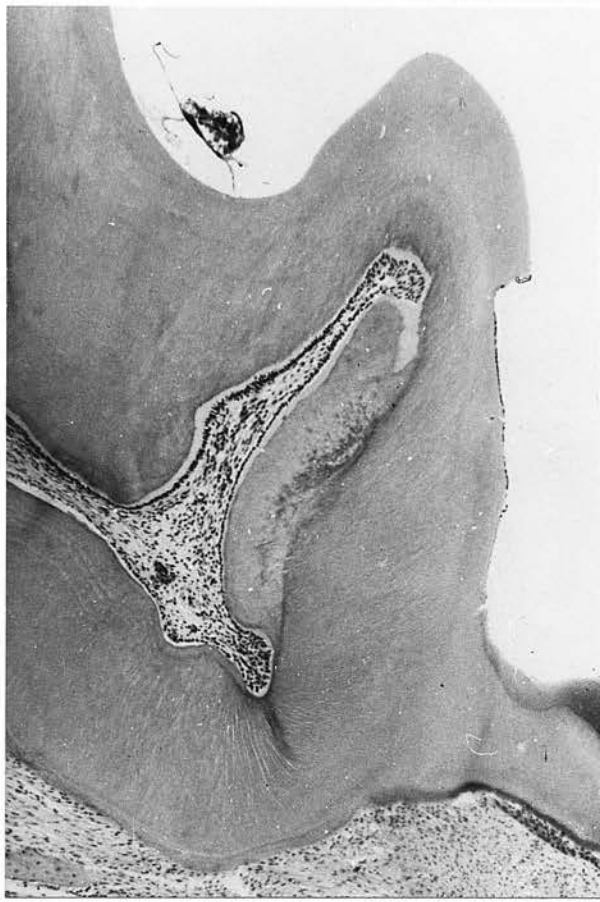
H. & E. x 48.

Fig³I2. Rat Molar with exposed pulp, the cavity filled with copper amalgam and examined after two months. A thick wall of dentine separates the vital pulp in the adjacent pulp horn from the extensive area of necrosis beneath the exposure.



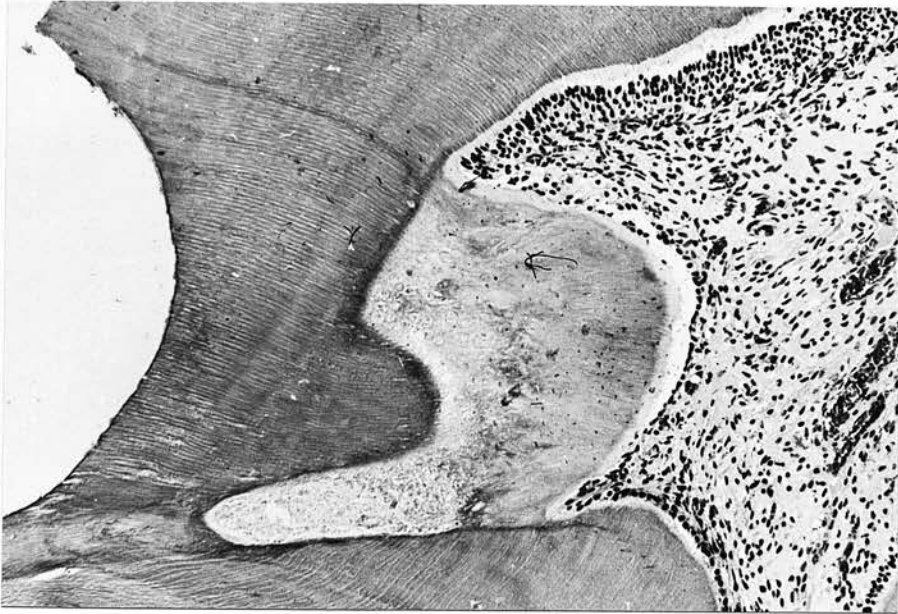
H. & E. x 60.

Fig³I3. Rat Molar with non-exposed pulp, the cavity filled with silver amalgam and examined after two months. A thick dentine bridge is present in the pulp horn adjacent to the cavity with a small area of necrotic pulp tissue above the bridge.



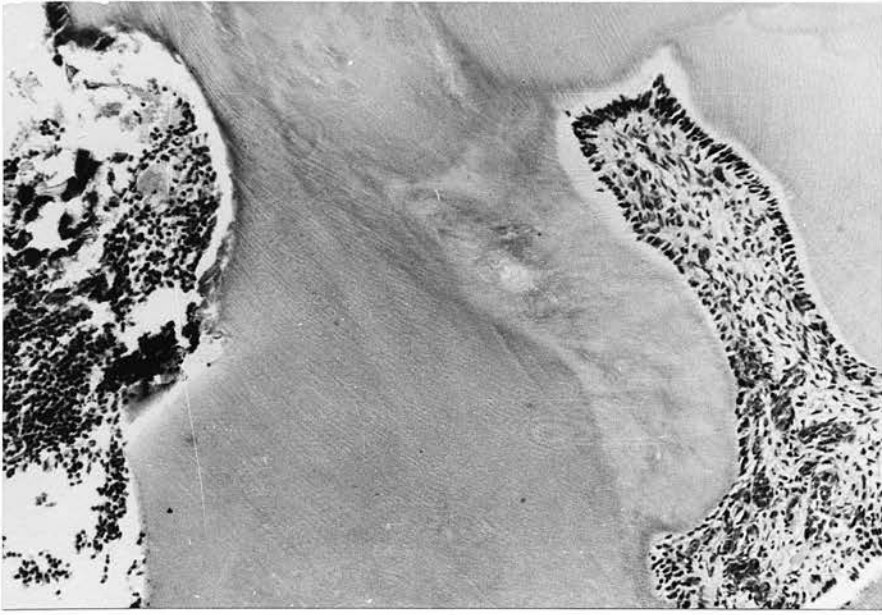
H. & E. x60.

Fig³T4. Rat Molar with unexposed cavity filled with copper amalgam and examined after three months. A well formed layer of secondary dentine has developed in relation to the cavity floor, which can be seen on the right.



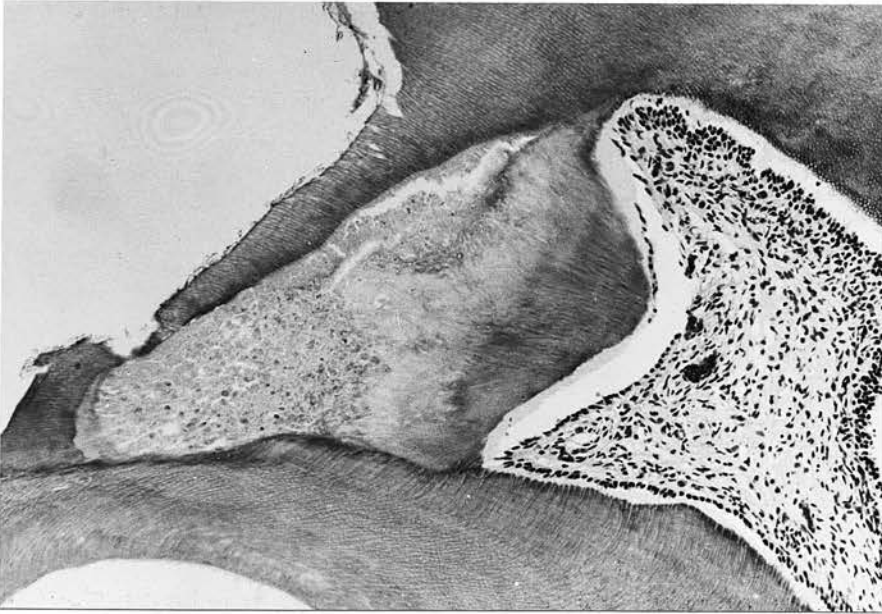
H. & E. x120.

Fig³T5. Rat Molar with exposed pulp, the cavity filled with copper amalgam and examined after three months. A well formed dentine bridge has developed beneath a small area of necrotic pulp tissue related to the exposure which was present in a serial section. Note the normal, vital pulp.



H. & E. x120.

Fig³I6. Rat Molar with unexposed cavity filled with copper amalgam and examined after two months. A layer of secondary dentine has formed in relation to the base of the cavity on the left which contains some inflammatory cells (probably from gingival inflammation).



H. & E. x120.

Fig³I7. Rat Molar with unexposed cavity filled with copper amalgam and examined after four months. A dentine bridge has formed beneath an area of pulp necrosis related to the base of the cavity on the left.

HISTOLOGICAL FINDINGS - RETAINED RESTORATIONS

1 MONTH

Of the teeth examined 1 month after placement of the restorations 2 (15%) of the cavities filled with copper amalgam and 2 (15%) of the cavities filled with silver amalgam were unexposed. Some other cavities were so shallow that no associated pulp reaction occurred and are therefore not included in the previous figures.

Of the two unexposed cavities filled with copper amalgam one showed evidence of walling and one showed bridging. The two cavities filled with silver amalgam showed evidence of dentine bridging. The pulp in the two teeth filled with copper amalgam showed that there was slight hyperaemia present at the time of sacrifice but the pulps of those filled with silver amalgam were normal.

Eleven cavities (85%) filled with copper amalgam and twelve (86%) filled with silver amalgam were exposed. The frequency of walling and bridging is given in TABLE 4 (A and B). The thickness of the dentine wall varied from very early dentine formation (Fig.3.7 and 8) to a layer of dentine of up to 80 micrometres in thickness (Fig.3.9, 10 and 11). Examination of the numbers shows that there was no correlation between the amount of dentine formed and the hyperaemia or inflammatory cell infiltration in the pulp tissue when the animals were sacrificed (Fig.3.11).

2 MONTHS

At 2 months 3 (16%) of the cavities filled with copper amalgam and 4 (23%) of the cavities filled with

silver amalgam were found to be unexposed. Secondary dentine or a dentine bridge had formed in all these examples (Fig.3.13 and 16).

Of the cavities filled with copper amalgam 10 (53%) were exposed and 8 (47%) filled with silver amalgam were exposed. The histological picture was similar to that exhibited at 1 month but on the whole the thickness of the dentine bridging and walling had increased by comparison with similar reactions seen in the 1 month group of teeth. The distribution of dentine bridging and walling was equal for both amalgams (Fig.3.12).

3 MONTHS

At 3 months 6 (26%) of the cavities filled with copper and 2 (20%) of the cavities filled with silver amalgam were unexposed. Again secondary dentine and dentine bridging were in evidence (Fig.3.14).

Of the cavities filled with copper amalgam 7 (30%) were exposed and of the silver amalgam 4 (40%) were exposed. The pulps in this category associated with both types of amalgam filling showed a greater tendency to walling (Fig.3.15) and there were 3 teeth associated with copper amalgam where necrosis of the radicular pulp tissue had taken place.

4 MONTHS

Since no silver amalgam restorations were retained at the end of this period no comparison with copper amalgam can be made. 2 (17%) of the copper restorations were unexposed, one exhibited secondary dentine, the other a dentine bridge (Fig.3.17). The 7 (58%) exposed pulps

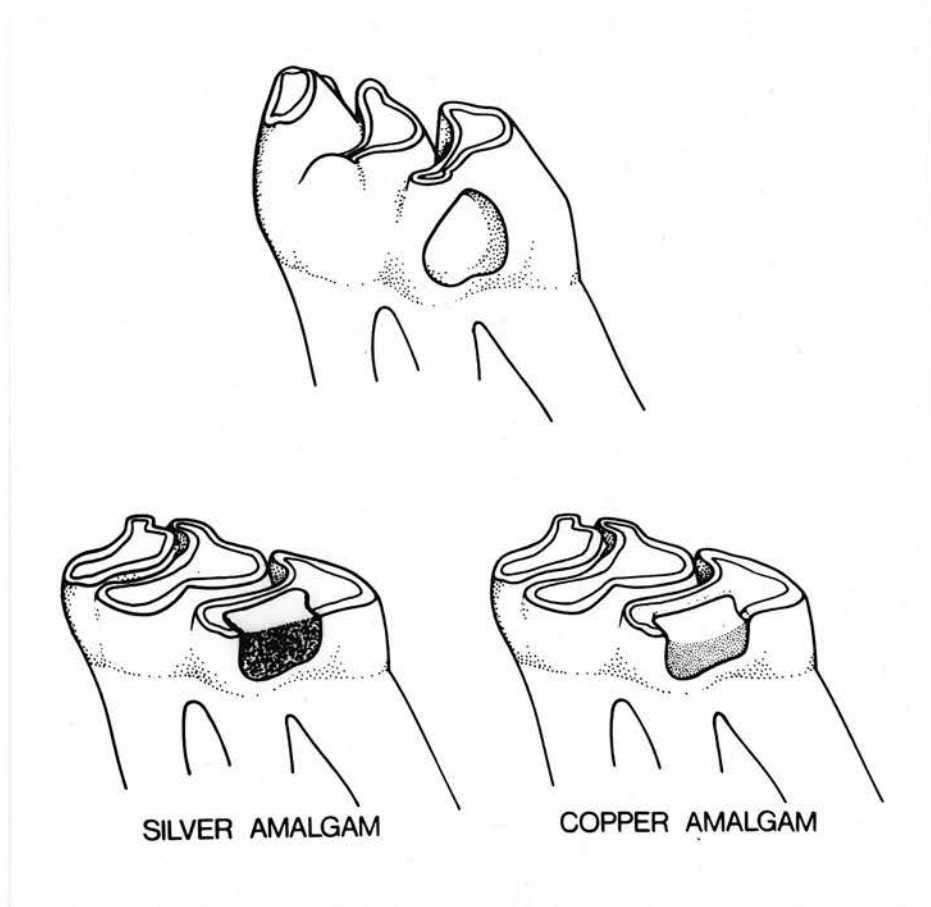
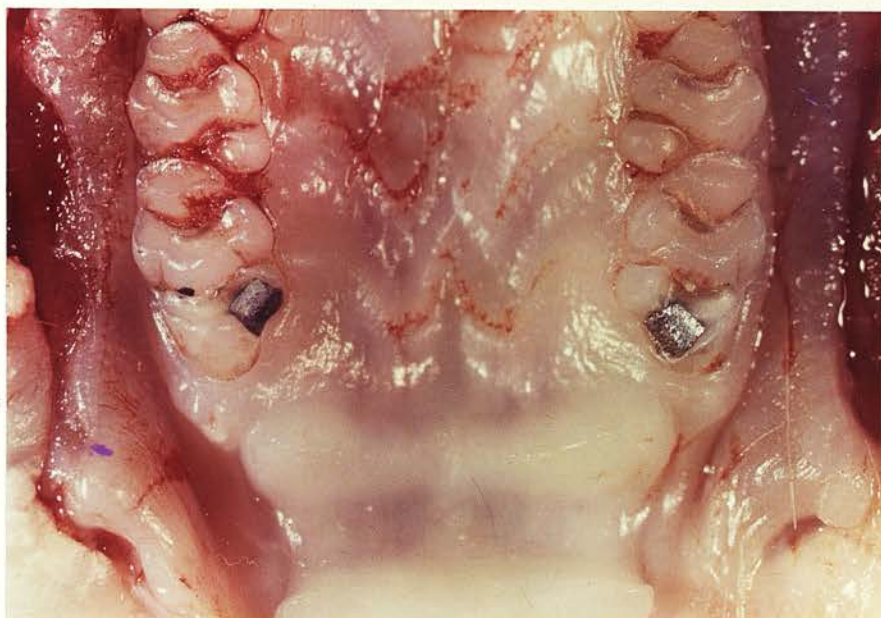


Fig. 18.

Sketches showing the results of attrition and a comparison of copper amalgam and silver amalgam restorations in the rat maxillary molar tooth three months after placement of the restorations.

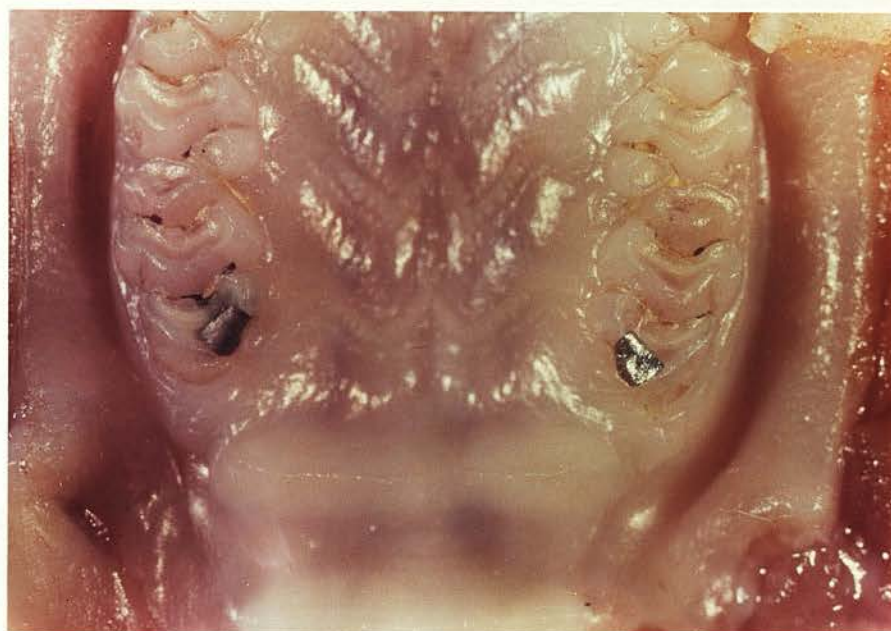


Fig³I9.

Copper Amalgam

3 months

Silver Amalgam



Fig³I9a.

Copper Amalgam

3 months

Silver Amalgam



Fig:20. Copper amalgam at three months.



Fig:20a. Silver amalgam at three months.

associated with copper amalgam restorations exhibited walling.

Discussion

The choice of laboratory animals as substitutes for clinical human material has always been controversial. The difference in scale is obviously important particularly in this study where the cavity size was diminished from the human situation by approximately ten times. The assumption, therefore, that the silver amalgam would exhibit an entirely similar performance to the human situation would be fallacious. The copper amalgam made from finely divided copper was placed at much less of a disadvantage in this respect although the situation in which both amalgams were used is identical.

The location of the cavity had an effect on the retention of the restorations especially after the 3 month period when, as a result of attrition, the cavity form changed (Fig. 3, 18, 19 and 20). Examination by stereomicroscope revealed that the copper amalgam restorations differed from the silver amalgams in that they exhibited a lower abrasion resistance than the surrounding occlusal surface of the tooth which, at 3 and 4 months is almost entirely dentine. The concavity or "cupping" of the copper amalgams produced by attrition, protected the restoration from dislodging forces. In contrast the silver amalgams abraded less quickly and remained higher than the occlusal surfaces of the teeth. The burnished margins and polished occlusal surfaces of the silver amalgam

restorations showed that they were in functional occlusion and therefore were more easily dislodged than the copper amalgams.

KAKEHASHI et al. (1965) showed that the prognosis for continued vitality of a surgically exposed dental pulp in the rat is poor unless bacteria are excluded from the site. In the present study there was no evidence to show that copper amalgam produced effects on the rat molar pulp which differed from those of silver apart from those changes which were directly attributable to loss or retention of the restoration. The exposures of the pulp were minimal in this study whereas those of previous studies ROWE (1967), KAKEHASHI (1965 and 1969) were, by intention, large.

CONCLUSIONS

1. On the evidence obtained from 175 amalgam restorations placed in the molar teeth of rats, there was no indication that an unlined copper amalgam restoration differed in its effects on the dental pulp from those of an unlined silver amalgam restoration under similar experimental conditions.
2. The fact that a copper amalgam restoration is likely to remain in place for a longer period of time was found to be an important factor in the maintenance of pulp vitality.

CHAPTER 4

TISSUE CULTURE STUDY

TISSUE CULTURE STUDIES

INTRODUCTION

The two constituents of copper amalgams copper and mercury are both toxic. Since few reports, (PREISSECKER, 1936), exist of serious systemic toxic effects arising from the use of copper amalgam as a restorative material, any such effects must be a matter of degree. Whether an amalgam can release quantities of its separate components or their compounds, which dissolve in saliva and are taken up in the tooth substance, depends on its solubility. Copper amalgam would appear to be more soluble than silver amalgam. The slow setting reaction described by CHAO and COSTA (1968), the continuous loss of weight found by FRASER (1929) in her bacteriological studies and, in addition, the characteristic green discolouration of teeth restored by copper amalgam is sufficient evidence of this.

Owing to the difficulty of evaluating the action of the material, in vivo, the aim of this study was to investigate the extent of ionic migration of copper and mercury from copper amalgam in a controlled fluid medium. It was decided that the effects on tissue culture growth could also be studied if a culture medium were used, (YAMAGAMI and KAWAHARA, 1970).

METHOD AND MATERIALS

Discs of pure silver and pure copper 6 mm in diameter and 2 mm thick were manufactured. Discs of identical dimensions were also made by packing freshly prepared copper amalgam and silver amalgam into special moulds. The silver discs and copper discs were polished and sterilised by autoclaving. By means of a stainless steel loop fastened to a glass slide a drop of mercury of similar size to the discs (6 mm diam.) was retained in position.

The following metals were, therefore, set up on 20 x 40 mm coverslips (glass slide for the mercury):

1. Copper
2. Copper amalgam (freshly prepared)
3. Silver
4. Silver amalgam (freshly prepared)
5. Mercury (+ Stainless steel wire)

A HEP 2 cell line was used in the experiment. The 5 coverslips were placed in 60 mm diameter Petri dishes which were seeded at 75,000 cells per cm^3 with 10 cm^3 Eagle's minimum essential medium. Controls were set up using sterilised glazed porcelain discs (6 mm diam.) and also untouched subbed coverslips. The cultures were gassed with 5% CO_2 and air and incubated at 37°C for 1, 2, 3 and four days. Each day 1 cm^3 of media was removed for analysis and replaced by 1 cm^3 of fresh media. After incubation the coverslips and slides were washed in phosphate buffered saline, fixed in

absolute ethanol and stained by haematoxylin and eosin.

The daily extractions of media were analysed by spectrophotometer for copper which could not be detected at less than 2 parts per million. Mercury analyses were carried out for the mercury by direct reduction with stannous chloride. Analysis for total mercury was carried out after overnight digestion with sulphuric acid and potassium permanganate followed by direct reduction with stannous chloride to break down any organically bound mercury. The mercury analyses were carried out to an accuracy of 0.01 p.p.m. by Dr. T. L. Coombs at the Institute of Marine Biology in Aberdeen.

Effects of tissue culture growth

- | | |
|-------------------|---|
| 1. Copper amalgam | - No live cells on any sample. |
| 2. Copper | - No growth at 4 days.
Scattered growth at 2 days. |
| 3. Silver amalgam | - Continued growth 15 mm from the disc. |
| 4. Silver | - Continued growth 4 mm from the disc. |
| 5. Mercury | - No live cells on any sample. |
| 6. Controls | - Normal growth. |

RESULTS

Analyses for Copper (n = 3)

parts per million

From Copper \bar{x} range

1 day	10.6	8.5 - 11.2
2 day	15.2	13 - 17.8
3 day	21.6	15.5 - 25.5
4 day	28.9	21.5 - 35

From Copper amalgam \bar{x} range

18.5	12.2 - 22.5
41.7	24.8 - 59.2
76.3	72 - 79
100.2	83.8 - 115

Analyses for Mercury (n = 3)

parts per million

From Mercury \bar{x} range

Total 4 day	0.23	0.12 - 0.41
Free Hg ⁺	0.05	0.00 - 0.08

From Copper amalgam \bar{x} range

0.72	0.57 - 0.82
0.33	0.00 - 0.08

From Silver amalgam \bar{x} range

Total 4 day	0.05	0.00 - 0.15
Free Hg ⁺	NONE DETECTED	

DISCUSSION

This study suggests that copper amalgam is a more effective source of copper and mercury than the free elements themselves. Analyses of the tissue fluids show that approximately 3 times the total mercury and copper are available from copper amalgam compared with similar specimens of pure mercury and copper.

Undoubtedly the copper amalgam will not be entirely homogeneous from the point of view of formation of the electron or intermetallic compound. There will be regions where "free copper" and "free mercury" are in intimate contact, and even although this may happen to a very small extent, the effect will be to form an electrolytic cell in which electrons will flow, and diffusion or migration of copper ions and mercury ions will occur.

The data for this study represents amounts accumulated in a static environment, in contrast to the situation in vivo. It is unlikely that the concentrations of either mercury or copper in this study would have any serious systemic effects, (FRYKHOLM, 1957 and 1969, and GOLDWATER, 1971).

CHAPTER 5

BACTERIOLOGICAL STUDY

A BACTERIOLOGICAL STUDY

The studies of MORTENSEN et al. (1965), BROWN & WHEATCROFT (1966), ELLIS & BROWN (1967) and ATTO et al. (1970) were attempts to relate microleakage to caries.

MORTENSEN et al. (1966) demonstrated that variations in temperature, such as would be experienced by the natural dentition, increased microleakage.

BROWN & WHEATCROFT (1966) demonstrated that diffusion of a nutrient factor through the tooth substance was necessary before natural carious lesions occurred in intact teeth and in those with an unfilled cavity.

ELLIS & BROWN (1967) studied the effects of a cavity liner by comparing lined and unlined restorations with regard to caries prevention. They found that lined restorations were less vulnerable to caries than were unlined restorations.

ATTO et al. (1970) compared silver amalgam with copper amalgam in a similar study and found more carious lesions associated with copper amalgam restorations than with silver amalgam.

This evidence suggests that caries can occur around restorations exhibiting marginal microleakage. The microleakage is increased by temperature variation and decreased by good adaptation and/or the use of a cavity liner. The effect of dimensional change is obscure but it would be logical to expect an amalgam that exhibits a setting contraction to exhibit more microleakage than one that had shown a setting expansion.

The aim of this study was to investigate any antibacterial action at the interface between tooth substance and copper amalgam and to compare this with observations on silver amalgam under identical experimental conditions. Since any effect that a restoration would have on the occurrence of caries would take place at the tooth/filling interface, reproduction of this situation is important. Direct access to the interface is also required to detect the presence of bacteria there.

It was decided to develop a test model that could take account of as many variables as possible. A flat tooth/filling interface offered advantages for surface finishing and consistent reproduction and so amalgam discs of uniform size placed against tooth sections formed the basis of this experiment. Refinements were introduced into the system as other variable factors were determined and procedures were devised to control them.

Thus, an early model in which a Petri dish was used, illustrated the problem of evaporation from large fluid surfaces and it was necessary to contain each experiment individually in smaller glass cylinders. This requirement not only imposed problems of access but also made it necessary to use materials that would withstand sterilisation and that could be in contact with culture fluids without exerting any toxic effect on the test organism.

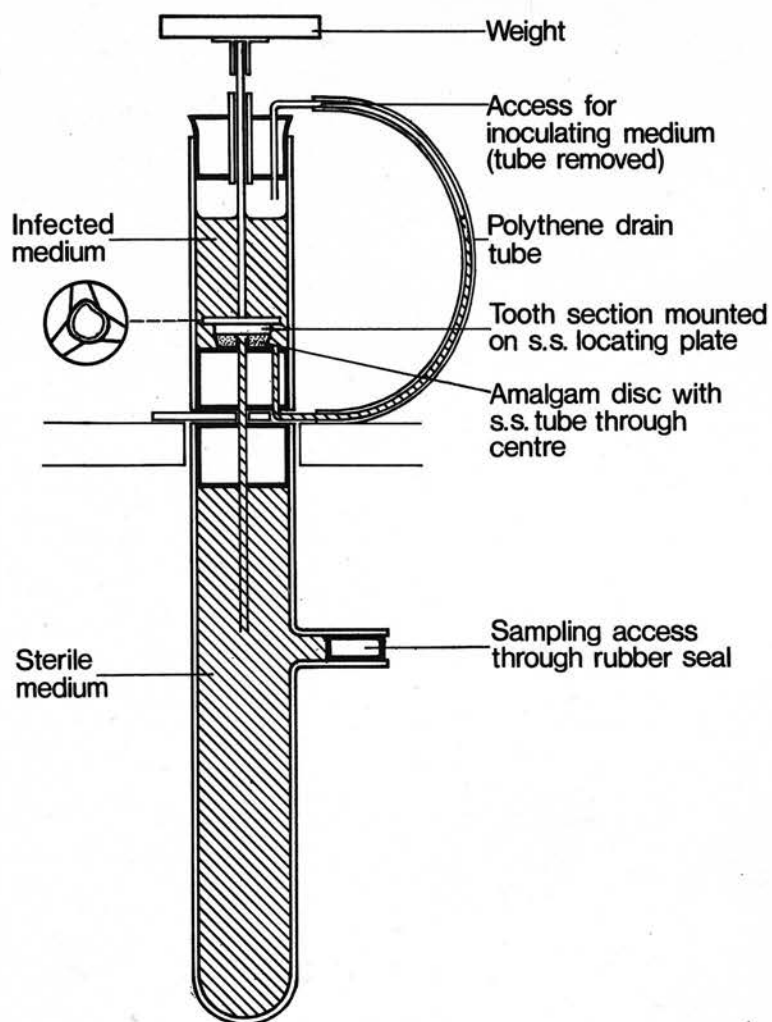


Fig. 5.I

A sectional drawing of the final assembly used in the bacteriological study.

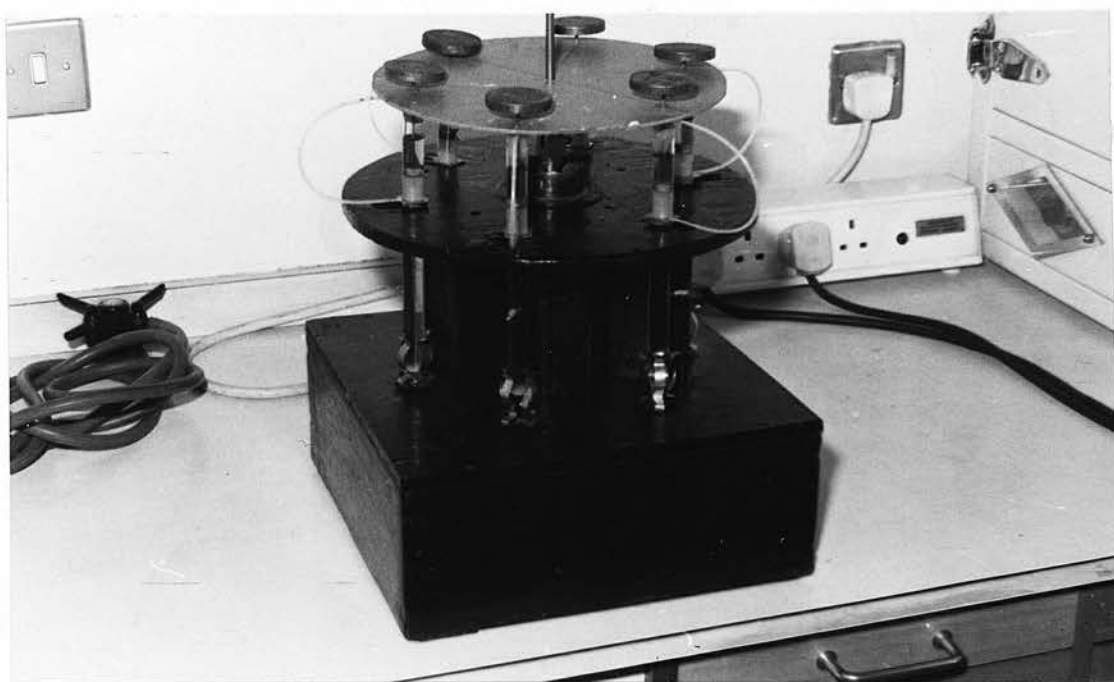


Fig. 5.2

The apparatus used to hold the six assemblies. The apparatus has been developed for future experiments to provide intermittent stress situations at the tooth/amalgam interfaces.

METHOD AND MATERIALS

Ethylene oxide gas was used for sterilisation. It was decided that sterility at the tooth/filling interface could not be guaranteed unless the two components could be separated for sterilisation. The amalgam discs (7.5 mm diam. and 3 mm thick) were fabricated by packing against a control surface of plate glass.

The tooth sections (coronal, 2 mm thick) were ground on one side with pumice on a plate glass slab. The surfaces were examined for flatness on a glass microscope slide.

The tooth sections were then cemented with an epoxy resin* to a stainless steel locating plate.

The apparatus was assembled for sterilisation as shown in Fig. 5.1, but without the weight. The amalgam discs were drawn down on to the lower stoppers by means of the stainless steel tubes and sealed on the underside with epoxy resin. The tooth slice and backing plate were introduced into the tube, the ground surface of the tooth slice opposing the top or control surface of the amalgam disc. The two components were held very loosely together during sterilisation by fixing the stainless steel rod, which would eventually support the weight, in position by means of masking tape. (During the experiment a weight of approximately 85 gm was placed on this rod, to ensure that the interface remained closed, eliminating the effects of dimensional change.)

The experiments were set up in groups of six as

* "Araldite" - Ciba-Geigy

shown in Fig. 5.2, three of copper amalgam and three silver amalgam.

- (1) The lower sterile test tube was filled with horse flesh digest medium (CRUICKSHANK, 1968). This fluid shows a characteristic cloudiness when contaminated by Streptococcus viridans. This was distinguished from cloudiness caused by the presence of other contaminants and discolouration caused by the presence of copper amalgam.
- (2) The top tube complete with the stopper was introduced into the lower test tube until fluid exuded from the interface. This ensured that there was uninterrupted fluid contact from the amalgam interface to the indicator medium in the lower test tube.
- (3) The weight was then applied via the stainless steel rod to a locating hole in the centre of the stainless steel backing plate which located the tooth slice over the centre of the amalgam.
- (4) The top tube was filled with sterile medium by removing the top connection of the polythene tube and injecting from a sterile syringe and needle into the exposed stainless steel tube.

- (5) The apparatus was placed in a 37°C incubator for 48 hr and checked for sterility.
- (6) The sterile medium in all the upper tubes was changed to avoid the accumulation of copper and mercury ions, which might affect the viability of the organism in those containing copper amalgam.
- (7) An inoculum of Streptococcus viridans was introduced into the top tube as in (4) above.
- (8) The experiment was examined twice in every 24 hr and concluded when the lower tube showed contamination or after 5 days. The fluid from the upper tube was then drained off and a small amount of the specimen was used to seed one half of a blood agar plate to check for the presence of viable Streptococcus viridans. A similar check was carried out by drawing off a small amount of the medium in the lower test tube using a sterile syringe and needle. This served to check that any growth in the lower test tube was caused by the inoculum and not by other contaminants.

In order to ensure that the experimental apparatus without the tooth/amalgam interface allowed bacteria to pass into the lower tube, 25 control experiments were carried out using the same apparatus as

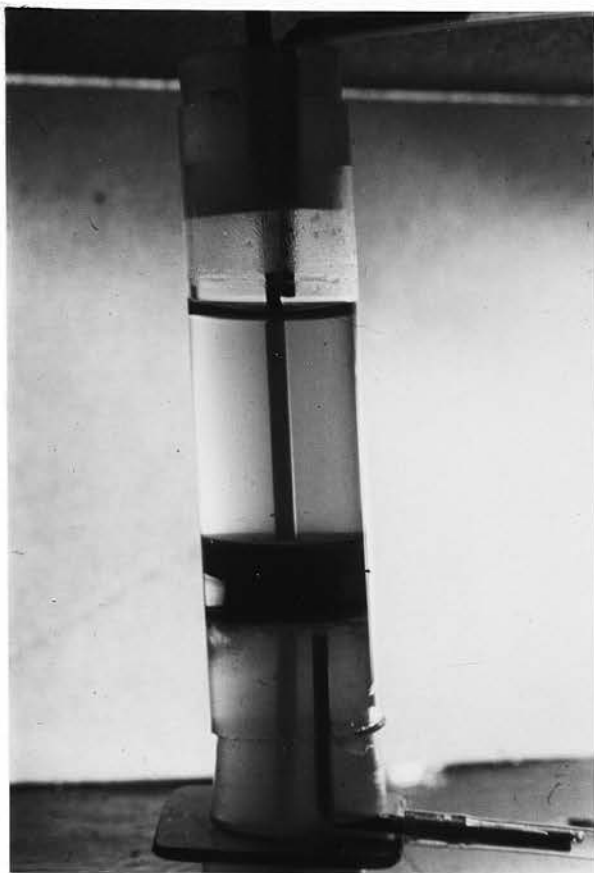


Fig. 5.3

An illustration of the top tube of an assembly containing an uncoated copper amalgam showing the green discolouration in the culture medium below the stainless steel locating plate.

illustrated in Fig. 5.1, but without the amalgam disc and tooth slice. In other words free access from the top to the bottom tube was arranged via the stainless steel tube. All 25 exhibited growth in the lower tube within 15 hr.

RESULTS

	Total number of experiments	Experiments which showed growth in the lower tube within 5 days
	Silver amalgam 26	22 (84.6%)
A.	Copper amalgam (uncoated) 26	7 (26.9%)
B.	Copper amalgam (coated) 19	9 (47.3%)

DISCUSSION

Results showed that there was a significant difference between the copper amalgam and the silver amalgam experiments. However it was assumed, since most of the top tubes containing the copper discs showed a difference in colour from those of silver (Fig. 5.3) after 24 hr, that the copper amalgam media contained a high concentration of copper ions. It was, therefore, possible that the viability of the bacterial challenge would be reduced by this time and a comparison valueless. The copper amalgam experiments were, therefore, repeated but this time two coats of cellulose acetate varnish were applied to the periphery of the copper amalgam discs and also the underside contacting the silicone rubber bung. Results of the second series of copper amalgams are shown in B above.

Analysis of the media in 18 top tubes and 18 bottom tubes for copper in the series B copper amalgams were carried out. The copper concentrations in the upper tubes containing copper amalgams varied between 10 - 40 parts per million. The silver amalgam experiments showed 1 - 2 p.p.m. of copper in the top tube. The surface area of the exposed part of the copper amalgam disc in series A, the uncoated specimens, was approximately 70 mm^2 and the contents of the top tube 2 - 3 cm^3 . This was a smaller area of copper amalgam exposed per cm^3 of fluid medium than that which SMITHERS & PARFITT (1955) claimed was necessary to produce a persistent bacteriocidal effect in a static environment.

It was decided to measure the pH in the top tubes of Series B to see if there was a relationship between the pH in a given tube at the end of an experiment and the copper content of the medium, but the pH value did not fall below 6 and no such relationship was found.

Statistical analysis of the study shows that the difference between copper amalgam and silver amalgam in the number of times growth appears in the lowest test tubes is highly significant, $p < .001$. ($t = 5.75$)

The difference between the modified copper amalgam (series B) and the silver amalgam is also significant but not so highly as in the previous situation. In this case $.01 > p > .001$. ($t = 2.28$)

The difference between the copper amalgams, series A and B, is not significant, $.20 > p > .10$ ($t = 1.99$).



Tooth
slice.

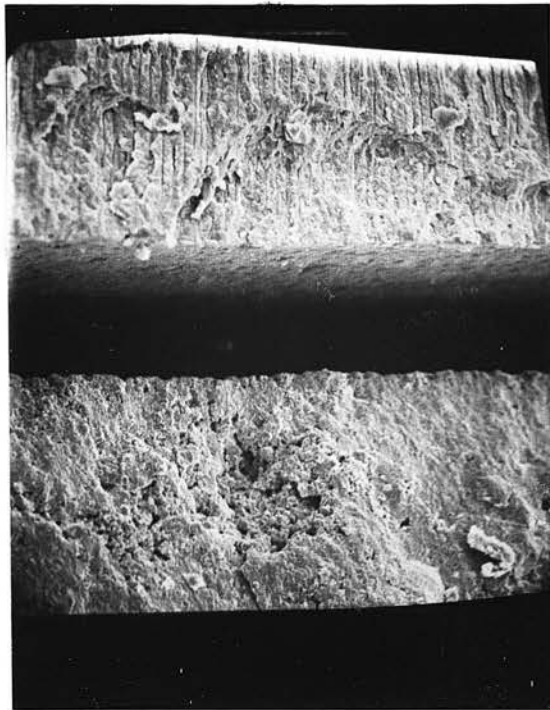
Silver
Amalgam.

SEM x65.

Fig. 5•4

Fracture section* of the interface between the silver amalgam and the tooth slice. The width of the space between the amalgam disc and tooth slice is 30 micrometres. (The overall sectional width of the amalgam and tooth slice related to this photograph would be 32 centimetres).

*see text.



Tooth
slice.

Copper
amalgam.

SEM x260.

Fig. 5.5

Fracture section of the interface between copper amalgam and the tooth slice. The width of the space is 60 micrometres. (The small sectional width of the amalgam and tooth slice related to this photograph would be 130 centimetres).



Tooth
Slice.

Copper
amalgam.

SEM x260.

Fig. 5.6

Fracture section of the interface
between copper amalgam and the tooth
slice. The width of the space is
100 micrometres.



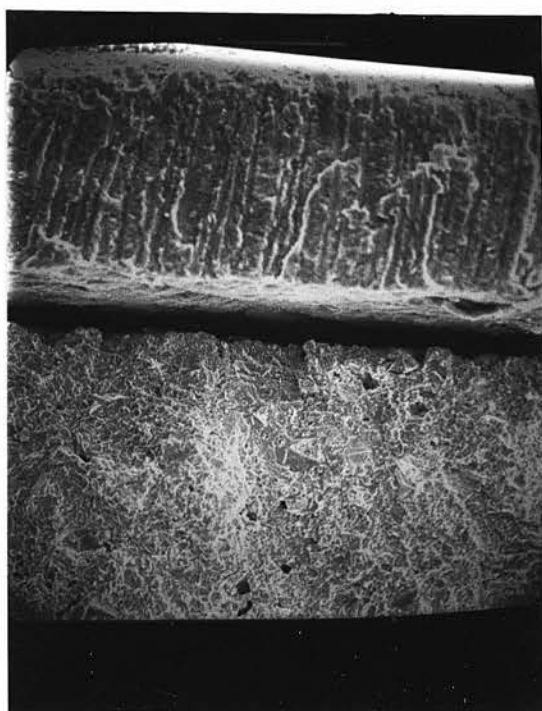
Tooth
slice.

Copper
amalgam.

SEM x1300.

Fig. 5.7.

Fracture section of the interface
between copper amalgam and the tooth
slice. The width of the space is
20 micrometres.



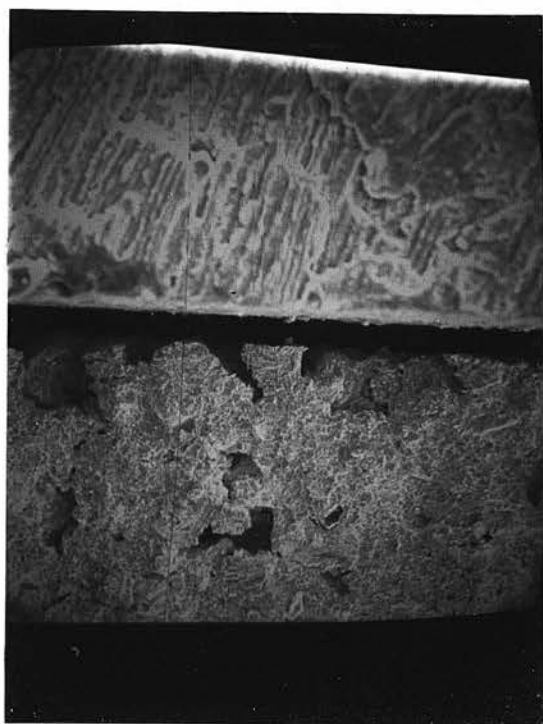
Tooth
slice.

Silver
amalgam.

SEM x260.

Fig. 5.8

Fracture section of the interface
between silver amalgam and the tooth
slice. The width of the space is
30 micrometres.



Tooth
slice.

Silver
amalgam.

SEM x260.

Fig. 5.9

Fracture section of the interface between silver amalgam and the tooth slice. The width of the space is 20 micrometres. The effect of the alloy particle size on the effective width can be seen.

At the end of each group of experiments, when the fluid medium had been drained for analysis, resin* was injected through the polythene drain tube shown in Fig. 5.1 so that the amalgam disc and tooth slice were held together at their edges when the resin had set. This unit was then sectioned using a high speed air turbine handpiece to cut down almost to the line of the interface; in amalgam on one side and in tooth substance on the other. Final separation was done by fracturing, the two opposing cuts serving as a guide so that the fracture planes of the amalgam and the tooth slice in the section were almost in alignment. The effects of any sectioning technique were thus avoided and the wide depth of focus of the scanning electron microscope (SEM) compensated adequately for any unevenness in the surfaces. (Fig. 5.4, etc.) Three copper amalgam and three silver amalgam specimens were examined by SEM. The spaces between the tooth slice and the amalgams varied from 20 - 115 micrometres in width with an average of about 40 micrometres. It was apparent that with both types of amalgam the width of the gap at the interface offered no obstruction to the size of the test organism (1 - 1.5 micrometres). This evidence obtained from the above studies seems to indicate that the presence of copper ions

* "Marco" Resin 5B 28c

may be a key factor in controlling the movement of bacteria. However, it is important to recognise the limitations of the experimental model at this stage. No attempt has been made to trace the spread of bacteria across the interface. It was accepted that the demonstration of bacterial access to the lower section of the model was sufficient proof that such a spread occurred. The control studies adequately support such a conclusion.

SUMMARY AND CONCLUSION

A model of the tooth/amalgam interface was designed which made it possible to eliminate or control the effects of:

- (a) The dimensional change of the amalgams on setting. The two components of the interface were flat and weighted in apposition.
- (b) Variations in cavity size. The cavity size in the model was represented by the area of the amalgam discs which were uniform in size.
- (c) The finish of the cavity walls or tooth surface. These were represented by the mating surface of the tooth slice in the model and were identically prepared by

grinding with pumice on a plate glass slab.

- (d) The surface against which the amalgams were packed was plate glass.

It was found from 71 experiments using the apparatus described above that a significantly higher number of experiments incorporating silver amalgam permitted bacteria to pass through the interface than the number using copper amalgam.

CHAPTER 2

INVESTIGATION OF TOOTH

CHAPTER 6

INVESTIGATION OF ERRORS

INVESTIGATION OF ERRORS

1. Heating the copper amalgam pellets

The actual temperature attained by the copper amalgam pellets was not checked against the thermometer reading. The presence of free mercury prevented the measurement of the temperature by thermistor. A rise in temperature not exceeding 5°C per minute was used throughout the study and when the pellets reached the required temperature the flask was shaken and reinserted in the heating element until the temperature was reached a second time. The minimum melting points used in this study were in agreement with those of HUTCHESON (1969) measured by differential thermocouples.

2. The mercury content of the copper amalgam specimens

Analysis of the mercury content of the copper amalgam as supplied was restricted to a few pellets from batches of each brand and it was possible that batches were not homogeneous. Thus the initial mercury content could not be assessed with absolute accuracy. With these limitations in mind the mercury content of the copper amalgams as supplied by the manufacturers was found by chemical analysis to be between 67 and 81 per cent (p.78) with an average of 72.4 per cent on four brands. (The average of 9 brands given by SKINNER (1946) was 66.8 per cent.)

It was found from an average of 20 mixes that 43 per cent by weight of the original copper amalgam was removed as excess mercury prior to and during condensation

(range 36 - 51 per cent). Because copper is soluble in mercury to only 0.003 per cent by weight (HANSEN, 1958) it was obvious that the excess contained very little copper and the percentage of residual mercury by weight in the copper amalgam specimens would be within a range of 50 - 60 per cent depending on (a) the brand and (b) the amount removed as excess. (GRANATH, 1971, gave 65.3 and 63.8 per cent.) A variation in mercury content from specimen to specimen was therefore expected as a result of (a) variations in the mercury content of the mass of copper amalgam from which excess mercury was squeezed, (b) differences in the amounts removed during condensation and (c) variation in condensation technique from specimen to specimen. For this reason an analysis of the crushed specimens was carried out and the mercury content was found to vary from 53.7 - 70.4 per cent. (See p.88)

3. Condensation Technique

A study of the technique required to adequately condense 40 copper amalgam specimens revealed that the number of thrusts of 35 - 44 N varied from 80 - 150. The reasons for this were as follows:-

1. The variation in mercury content between batches of the same brand and between different brands.
2. The variable amount of mercury released when a given weight of copper amalgam is heated to a given temperature. This is probably related to (1) above.

3. Condensation of copper amalgam at 35 - 44 N with a 2 mm diam. condensing instrument always produced excess mercury (or liquid amalgam) and even if this was constantly removed as packing continued, mercury or liquid amalgam always appeared on the surface of the specimen as extrusion pressure was applied and "soaked" into the specimen when the pressure was relieved.

To elucidate the possible effect of the variations in condensation technique, two series of specimens were packed varying the number of condenser thrusts from 25 to 200 in steps of 25 (Table 1).

No. of thrusts 35 - 44 N	Crushing Strength N/mm ²	
25	151.3	
50	132.0	159.9
75	136.1	190.4
100	219.4	205.6
125	213.3	208.2
150	213.3	216.3
175	142.2	195.5
200	175.2	

It can be seen that there was no ordered relationship between the number of condenser thrusts and the compressive strength of the specimens. The strength at 100 - 150

thrusts was not altered significantly by further condensation. 100 - 150 thrusts of 35 - 44 N was therefore chosen as suitable for the production of satisfactory specimens.

In an effort to eliminate the effects of variations in temperature of preparation and variations in condensation technique, 32 specimens of copper amalgam were prepared from pellets heated to 160°C and were packed using 100 thrusts per specimen. These were crushed after 7 days. The results shown on p.88 are repeated below:-

Compressive Strength at 7 days - N/mm^2				
Temperature of Preparation	n=	\bar{x}	s	
160°C	32	158.6	40.3	$v = 25.4\%$

It can be seen that there was considerable variation in the strength of the specimens. This was attributed to porosity in the specimens which because of the nature of the material occurred in a random manner and could not be avoided (JØRGENSEN et al., 1966).

4. Dimensional Change

(a) Instrument Error

An attempt was made, previously described on p.82, to eliminate the effects of creep in the "Mitronic" stand and the effects of temperature at the measuring head, but in control tests using the specimen die as a slip gauge an error of 1 - 1.5 micrometres was still present and could not be eliminated.

(b) Errors arising because of the nature of the material

- (i) Mechanical compression of the copper amalgam specimen under the 3 gm stylus weight might have increased the apparent contraction but, as the compression took place before the commencement of measurement the error from this source was accepted. It seemed reasonable to expect that it would be similar for all copper amalgam specimens tested.
- (ii) The extended setting time and soft texture of copper amalgam may permit flow to take place before the amalgam sets. Flow in the early stages of setting without any external force on the test cylinder would have increased the apparent contraction as in b (i). As this factor operated on all the copper amalgam specimens tested and could not be eliminated, errors from this source were also accepted.

5. Cavity Preparation in the rat molar study

The cavities in the rat molar teeth were prepared by holding the dental handpiece in the hand diagonally opposite to the tooth undergoing cavity preparation. Since the operator was right handed it is possible that there was a difference in the precision with which right

and left hand cavities were cut, but no obvious differences between right and left cavities were detected in the histological preparations and it was therefore assumed that the errors from this source were sufficiently small to be ignored. There were however differences in depth of over half the remaining dentine thickness, measured from the deepest part of the cavity using serial sections, which were not related to any side and which could not be eliminated.

6. The choice of silver amalgam in the rat molar study

New True Dentalloy (S.S. White) was used in this as it is a popular and representative dental silver amalgam. However, it exhibits a final setting expansion as do most silver amalgam alloys. It is obvious that comparative results would vary with different silver amalgams.

7. The tooth-amalgam interface in the bacteriological study

Sterilisation in ethylene oxide for 16 - 20 hr was followed by a clearing period of 5 days in dry conditions. The tooth slices would have dried out in this time and it is possible that, despite the stainless steel backing plate, some distortion could have occurred and disturbed the even contact between the amalgam discs and the tooth slices. This was not evident on visual inspection before the introduction of the culture medium. It was recognised, however, that small errors could have escaped detection but they were accepted as they would occur at random and operate equally on both groups of specimens tested.

SUMMARY AND CONCLUSIONS

SUMMARY AND CONCLUSIONS

1. The relevant literature on copper amalgam has been reviewed and includes a survey of research techniques used to evaluate the efficiency of dental restorations in vitro and in vivo.
2. Copper amalgam was compared to silver amalgam because they are materials which perform a similar function in restorative dentistry. Both are used in children's dentistry. Chemically and structurally, however, the two materials differ considerably and there are differences in their preparation and handling for use in the mouth.
3. Metallurgical examination of copper amalgam specimens by incident light and electron probe analysis showed areas almost entirely composed of copper which indicated that the distribution of copper in any given sample was uneven. It is possible that this characteristic of copper amalgam, together with random porosity of the specimens could account for the inconsistent results of some of the mechanical tests.
4. In an investigation of the mechanical properties of copper amalgam over 100 specimens were tested. The following average values were obtained:-

Property	No. of Specimens	Av. Value
Dimensional change on setting	61	= 0.123% contraction
Compressive strength (24 hr)	65	= 120 N/mm ²
Compressive Strength (7 day)	32	= 159 N/mm ²
Tensile strength (24 hr)	29	= 17 N/mm ²
Vickers Hardness (24 hr)	20	= 96

5. From a study of the dimensional change on setting it was concluded that copper amalgam always exhibits a setting contraction. However, no relationship could be found between the temperature of preparations of the copper amalgam pellets and the dimensional change on setting as found by WORMINGTON (1939).
6. By comparison with the initial setting time of a popular dental silver amalgam it was found that the setting of copper amalgam was undesirably slow.
7. Results indicated that the compressive strength and tensile strength of copper amalgam were extremely variable and were not related to the residual mercury content of the specimens. This may have been due to the difficulty of controlling all the variables in the production of test cylinders of copper amalgam and it is obvious that further work is necessary to elucidate the effect of porosity on these properties.

The average value for 24 hr compressive strength obtained in this study is approximately half of that obtained by TAYLOR (1929) but since he produced only three results the average figure of 120 N/mm^2 for 65 specimens is more reliable.

8. It was found that copper amalgam adapted to the walls of special moulds better than 5 types of dental silver amalgams used in this study. The difference in structure between copper amalgam and dental silver amalgam accounted for the differences in adaptation. This was clearly shown by scanning electron microscope studies.

9. Radioactive isotopes and dye techniques were used in a study of microleakage in 10 copper amalgam and 10 silver amalgam restorations in extracted teeth. Both copper amalgam and silver amalgam restorations exhibited microleakage at the margins. There was no evidence that the contrasting dimensional changes on setting of the two amalgams had any effect on the amount of microleakage.
10. Over 80 copper amalgam restorations and 80 silver amalgam restorations were placed in the molar teeth of rats. Histological sections of the teeth showed no significant difference between the effects of unlined copper amalgam and unlined silver amalgam restorations.
11. Concave attrition of the copper amalgam restorations protected them from accidental dislodgement from the teeth. Attrition of the silver amalgam was less evident and thus the restorations were more easily dislodged. This finding is contrary to what might be expected from materials of similar hardness (TWEEDALE, 1964) and might possibly be explained by a lower corrosion resistance and dissolution of the copper amalgam (FRASER, 1929).

In the study of the mechanical properties the Vickers Hardness of copper amalgam was found to be 96 and differed only slightly from that of the silver amalgam (98) examined for comparison.

14. There was an apparent contradiction between the findings of the study of the effects of the amalgams on the dental pulp in rat molar teeth and the toxic effects of copper amalgam on cells in tissue culture. It seems reasonable to suppose that the toxic effects observed in vitro were due to the comparatively static and sensitive tissue culture environment. In vivo the constantly changing environment backed up by the normal tissue reaction prevented the copper ions from exerting a harmful effect.
15. An experimental model of the tooth/amalgam interface was designed to study the movement of bacteria in this area. By comparison with previous studies of this type the number of variables was reduced. It was found from 71 experiments using this model that a significantly higher number of experiments incorporating silver amalgam permitted bacteria to pass through the interface than the number with copper amalgam.

ACKNOWLEDGEMENTS

I have to thank Professor D. M. Watt of the Department of Restorative Dentistry, Edinburgh University and Dr. R. C. Howie, Department of Chemistry, Heriot-Watt University, who supervised this study.

I have also to thank Professor L. E. Granath for an instructive visit to the Royal Dental School, Malmo, in 1970.

I am indebted to Dr. J. G. Collee of the Department of Bacteriology and Dr. D. M. W. Anderson of the Department of Chemistry, Edinburgh University, for their special advice and assistance.

I have to acknowledge the work of Dr. T. L. Coombs of the Institute of Marine Biochemistry, Aberdeen; Dr. D. Purves of the Edinburgh School of Agriculture and Dr. D. M. W. Anderson who carried out various copper and mercury determinations for this study.

I am also indebted to my colleagues who have helped me in a number of ways:- Professor G. S. Beagrie, Dr. C. E. Chapman, Dr. P. W. Ross, Dr. J. C. Southam, G. H. Bolas, J. S. Clyde, J. A. Hargreaves, M. W. Miller, G. C. Millar and G. H. Moody.

I am especially indebted to Mr. J. M. Mitchell and Mr. J. R. Turnbull for their technical assistance, Mr. A. Hunter, Miss J. Mitchell, Mr. R. Renton, Mr. I. Goddard, who prepared the illustrations and Mrs. Y. Blair who typed the manuscript.

BIBLIOGRAPHY

- 1961 An Examination of Dental
Corporations in the
United States - King
McGraw-Hill Book Co.
- 1963 Cavity Filling. Brill
Leiden, The Netherlands
1963
- 1971 & 72 Guide to Dental Materials
and Devices. 3rd. ed. Brill
Leiden, The Netherlands
- 1970 An X-ray study of denture
base material. Brill
Leiden, The Netherlands
1970
- 1971 Dental and denture
materials. Brill
Leiden, The Netherlands
1971
- 1971 The influence of the
dental alloy on the denture
base material. Brill
Leiden, The Netherlands
1971

BIBLIOGRAPHY

- 1970 The influence of denture
base material on the denture
base material. Brill
Leiden, The Netherlands
1970
- 1971 A study of the denture
base material. Brill
Leiden, The Netherlands
1971
- 1971 Study of denture base
material. Brill
Leiden, The Netherlands
1971
- 1971 The influence of the
denture base material on the
denture base material. Brill
Leiden, The Netherlands
1971
- 1971 Study of the denture
base material. Brill
Leiden, The Netherlands
1971
- 1971 Study of the denture
base material. Brill
Leiden, The Netherlands
1971

BIBLIOGRAPHY

- ALLAN, D. N. 1957 An Evaluation of Dental Conservative Restorations Thesis - King's, Newcastle-Upon-Tyne.
- ALLAN, D. N. 1968 Cavity Finishing. Brit. Dent. J. 125: 540-545.
- American Dental Association 1970 - 71 Guide to Dental Materials and Devices, 5th ed. p.22.
- ATTO, A. Y. et al. 1970 In vitro study of carious lesions around dental restorations. J. dent. Res. 49: 756-759.
- BALDWIN, H. 1897 Cement and amalgam fillings. Brit. Dent. Ass. J. 18: 255-257.
- BARBER, T. K. et al. 1964 The effectiveness of copal ether varnish under amalgam restorations. J. Prost. Dent. 14: 533-536.
- BARR, J. H. & GRESHAM, A. H. 1950 The detection of carious lesions on the proximal surfaces of teeth. J. Amer. dent. Ass. 41: 198-204.
- BASKER, R. M. & WILSON, H. J. 1969 A machine for packing amalgam specimens under simulated clinical conditions. Brit. dent. J. 126: 209-215.
- BASKER, R. M. & WILSON, H. J. 1971 Mechanical amalgam packers. Brit. dent. J. 130: 147-153
- BERENBAUM, R. & BRODIE, I. 1959 The measurement of the tensile strength of brittle materials. Brit. J. appl. Phys. 10: 281-287.
- BERGVALL, O. & BRANNSTROM, M. 1971 Measurements of the space between composite resin fillings and the cavity walls. Sven. Tandlak. Tidsk. 64: 217-226.
- BERMAN, D. S. & MASSLER, M. 1958 Experimental pulpotomies in rat molars. J. dent. Res. 37: 229-242.

- BRAIN, E. B. 1966 The Preparation of Decalcified Sections.
Thomas - Springfield.
- BRANNSTROM, M. & SOREMARK, R. 1962 The penetration of Na²² ions round amalgam restorations with and without cavity varnish. Odont. Revy. 13: 331-336.
- BRANNSTROM, M. & NYBORG, H. 1971 The presence of bacteria in cavities filled with silicate cement and composite resin materials. Sven. Tandlak. Tidskr. 64: 149-155.
- British Standards Institution 1961 Specification for Dental Amalgam Alloy, B.S. 2938
- British Standards Institution 1961 A Method for Vickers Hardness Test. B.S. 427: Part 1 Testing of Metals.
- BROWNLIE, J. R. 1875 Some properties of amalgams - shrinkage. Brit. J. dent. Sci. 18: 245-257.
- BROWN, L. R. et al. 1962 The diffusion of niacin through extracted human teeth and its effect on bacterial penetration into dentin. J. dent. Res. 41: 684-694.
- BROWN, L. R. & WHEATCROFT, M. G. 1966 Effect of the diffusion of microbial growth factors through tooth substance on the production of carious lesions in vitro. J. dent. Res. 45: 830-837.
- BORINSKY, P. 1931 The source of mercury in urine. Zahnarztl. Rdsch. 40: 221-229 (Ger)
- BOYDE A. & KNIGHT, P. J. 1969 The use of scanning electron microscopy in clinical dental research. Brit. dent. J. 127: 313-322.
- BOWDEN, S. T. 1950 The Phase Rule and Phase Reactions
Macmillan - London

- CHAO, F. & COSTA, M. 1968 The relationship between the potential of the copper amalgam electrode, its strength and structure. Bull. Soc. Chim. Fr. 2: 549-555 (Fr)
- COGGAN, J. G. 1932 Unpublished data, see WILSON 1933.
- COMBE, E. C. 1972 Notes on Dental Materials Churchill Livingstone - Edinburgh.
- CRUICKSHANK, R. 1968 Medical Microbiology pp. 744-745 Livingstone - Edinburgh.
- DACHI, S. F. & STIGERS, R. W. 1967 Reduction of pulpal inflammation and thermal sensitivity in amalgam restored teeth treated with copal ether varnish. J. Amer. dent. Ass. 74: 1281-1285.
- DEBRAY 1856 see HENLEY
- DE MAAR, F. E. 1968 Who introduced silver amalgam into dentistry? Nederl. T. Tandheelk 75: 313-321 and 395-404 (Dut)
- DIXON, C. & RICKERT, U. 1933 Tissue tolerance to foreign materials. J. Amer. dent. Ass. 20: 1458-1472.
- DOW, R. H. 1963 Mouldable amalgams for cast patterns. Brit: 924426.
- EAMES, W. B. 1959 Preparation and condensation of amalgam with a low Hg: alloy ratio. J. Amer. dent. Ass. 58: 78-83.
- EAMES, W. B. 1967 Factors influencing the marginal adaptation of amalgam. J. Amer. dent. Ass. 75: 629-637.

- EASLICK, K. A. 1935 Filling materials for children's teeth. J. Amer. dent. Ass. 22: 1050-1056.
- EASTON, G. S. 1941 Causes and prevention of amalgam failures. J. Amer. dent. Ass., 28: 392-400.
- EDEN, G. T. & WATERSTRAAT, R. M. 1967 Effects of packing pressures on the properties of spherical particle alloy amalgams. J. Amer. dent. Ass. 74: 1024-1029.
- EFFINGER, A. 1954 Histological examination of copper oxide cement and copper amalgam implants. D. Zahnarztl. Z., 9: 1289-1297 (Ger)
- EFFINGER, A. 1957 Experimental results of the mode of action of copper containing fillings. D. Zahnarztl. Z. 12: 1645-1651 (Ger)
- ELLIS, V. M. & BROWN, L. R. 1967 Application of an in vitro cariogenic technic to study the development of carious lesions around dental restorations. J. Dent. Res., 46: 403-408.
- EMELEUS, H. J. & ANDERSON, J. S. 1938 Modern Aspects of Inorganic Chemistry Routledge, London.
- FARRIS, E. J. & GRIFFITH, J. Q. 1949 The Rat in Laboratory Investigation Hafner, New York, 2 ed.
- FLAGG, J. F. 1891 Plastics and Plastic Filling Fagan & Sherman, Philadelphia.
- FLETCHER, T. 1876 Tube packing test for amalgams. Trans. odont. Soc. 8: 83-88, 90-92.

- 148
- FLETCHER, T. 1895 On copper amalgam.
Trans. Odont Soc. 7: p.91.
- FORSTEN, L. 1969 Physical Properties of
Dental Amalgams
Thesis - Turku.
- FRASER, J. 1929 A study of the efficiency
of dental fillings.
J. dent. Res. 9: 507-517.
- FRYKHOLM, K. O. &
ODEBLAD, E. 1955 Studies on the penetration
of mercury through the
dental hard tissues using
Hg 203 in silver amalgam
fillings. Acta. odont.
Scand. 13: 157-165.
- FRYKHOLM, K. O. 1957 Mercury from dental
amalgam: its toxic and
allergic effects.
Acta. odont. Scand.:
15: Supplement 22.
- FRYKHOLM, K. O. et al. 1969 Allergy to copper derived
from dental alloys as a
possible cause of oral
lesions of lichen planus.
Acta. dermat. venereol.
49: 268-281.
- FUSAYAMA, T. et al. 1964 Early and transversal
measurement of amalgam
expansion. J. dent.
Res., 43: 194-205.
- FUSAYAMA, T. 1967 Surface roughness of
amalgam fillings.
J. dent. Res., 46:
1019-1021.
- GOING, R. E. et al. 1960a Marginal penetration of
dental restorations as
studied by crystal violet
dye and I¹³¹. J. Amer.
Dent. Ass. 61: 285-300.
- GOING, R. E. et al. 1960b Marginal penetration of
dental restorations by
different radioactive
isotopes. J. dent. Res. 39:
273-284.
- GOING, R. E. &
MASSLER, M. 1961 Influence of cavity
liners under amalgam
restorations on penetra-
tion by radioactive
isotopes. J. Prosthet.
Dent. 11, 298-312.

- 149
- GOLDWATER, L. J. 1971 Mercury in the environment. Scientific American. 224: 15-21.
- GRANATH, L. E. 1961 Hardness of copper and silver amalgam. Odonto. Revy. 12: 368-376.
- GRANATH, L. E. & HAKANSSON-HOLMA, B. 1961 The occurrence of certain defects in copper amalgam restorations in the primary dentition. Odonto. Revy 12: 272-289.
- GRANATH, L. E. 1967 Studies on microleakage with restorative materials. I. Introductory experiments on amalgam. J. dent. Res. 46: 1131-1336.
- GRANATH, L. E. & SVENSSON, A. 1970 Studies on microleakage with restorative materials - A new air pressure method. Scand. J. Dent. Res. 78: 353-366.
- GRANATH, L. E. 1971 Studies on microleakage with restorative materials. Acta. Odont. Scand. 29: 65-73.
- GRANATH, L. E. & MÖLLER, B. 1971 Reaction of the human dental pulp to silver amalgam restorations. Acta. Odont. Scand. 29: 165-172.
- GUNZ, A. & DE GRIEFT. 1912 Copper amalgam. Compt. Rendu. 154: 357-358 (Fr)
- GURLEY, W. B. & VAN HUYSSEN, G. 1937 Histologic changes in teeth due to plastic filling materials. J. Amer. dent. Ass. 24: 1086-1816.
- HANSEN, M. 1958 Constitution of Binary Alloys. McGraw Hill, New York.

- HATT, S. D. 1959 The relationship of amalgam to the cavity wall. Dent. Pract. 10: 76-86.
- HARRIS, R. S. 1968 Art and Science of Dental Caries Research. Academic Press - New York and London.
- HEALEY, H. & PHILLIPS, R. W. 1949 A clinical study of amalgam failures. J. Dent. Res. 28: 439-446.
- HENLEY, N. W. 1965 Henley's 20th century Book of Formulas, Processes and Trade Secrets. Books - New York.
- HOFFMAN, M. M. & SCHOUR, I. 1940 Quantitative studies in the development of the rat molar. Anat. Rec. 78: 233-251.
- HOGEBOM, F. E. & HURST, I. L. 1923 Bacterial inhibiting powers exhibited by copper amalgam used for deciduous restorations. Pacific dent. Gaz., 31: 515-519.
- HOHN, H. 1948 Amalgam metallurgy. Wien Chem-Ztg. 49: 15-31 (Ger) (Chem. abs. 44: 4841e)
- HUME-ROTHERY, W. 1948 Atomic Theory for students of metallurgy Institute of Metals (Monograph and Report Series No. 3) - London.
- HUME-ROTHERY, W. 1955 Electrons, Atoms, Metals and alloys 2nd ed. Iliffe - London.
- HUME-ROTHERY, W. et al. 1951 The factors affecting the formation of 21/23 electron compounds in alloys of copper and silver. J. Inst. Metals. 80: 609-616.
- 1952

- HUSBAND, F. C. 1928 Copper amalgam as a root-filling material. J. Amer. dent. Ass. 15: 248-251.
- HUSBAND, F. C. 1932 A process for the production of copper amalgam from ingredients in aqueous solution. J. Dent. Res. 12: 321-326.
- HUTCHESON, D. 1969 Undergraduate Dissertation Heriot-Watt, Edinburgh.
- IRVINE, N. M. & RUSSELL, A. S. 1932 The solubilities of copper, manganese and some sparingly soluble metals in mercury. J. Chem. Soc. 1932: 891-898.
- JORGENSEN, K. D. 1965 The adaptability of dental amalgam. Acta. Odont. Scand. 23: 257-270.
- JORGENSEN, K. D. et al. 1966 The effect of porosity and mercury content upon the strength of silver amalgam. Acta. Odont. Scand. 24: 535-553.
- JORGENSEN, K. D. 1967 A structure study of amalgam III. Acta Odont. Scand. 25: 85-104.
- KAKEHASHI, S. et al. 1965 The effects of surgical exposures of dental pulps in germ-free and conventional laboratory rats. Oral Surg. 20: 340-349.
- KAKEHASHI, S. 1969 The exposed germ free pulp: Effects of topical corticosteriod medication and restoration. Oral. Surg. 27: 60-67.
- KANAI, S. 1966 Structure studies of amalgam II: the effect of burnishing the margins of occlusal amalgam fillings. Acta. Odont. Scand. 24: 47-53.

- KAWAHARA, H. et al. 1965 Biological test of dental and surgical materials by means of tissue culture. Proc. 2nd Conf. Int. Acad. Oral path. pp. 79-92.
- KNAPPWOST, A. 1951 Microleakage from plastic fillings is a factor in secondary caries. Dtsch. Zahnartzl. Z. 6: 602-609. (Ger)
- KORAN, A. & ASGAR, K. 1967 A comparison of dental amalgams made from spherical alloy and from a comminuted alloy. J. Amer. dent. Ass. 75: 912-917.
- KOSLOV, M. & MASSLER, M. 1960 Histologic effects of various drugs on amputated pulps of rat molars. Oral Surg., 13: 455-469.
- KOSLOWSKI, M. T. 1956 Mercury and amalgams in electrochemical analysis. Alma-Ata: Izdatel Acad. Nauk Kazakh. S.S.R. r11, K.80: 185 (Russ.)
- KRAUS, A. 1945 From engineering to physiology. Brit. dent. J. 78: 230-237.
- KRONFELD, R. 1933 Histopathology of the teeth and their surrounding structures Lea and Febiger - Philadelphia.
- LAVIS, J. F. 1954 Copper amalgam. Aust. J. den. 58: 321-322.
- LIHL, F. 1953 Amalgams of manganese, iron, cobalt, nickel and copper. Z. Metalkunde. 44: 160-166 (Ger)
- LIND, V. et al. 1964 Contact caries in connection with silver amalgam, copper amalgam and silicate fillings. Acta. Odont. Scand. 22: 333-341.

- LINDAHL, T. et al. 1968 Rhombohedrally distorted gamma phases in the copper-mercury and chromium-aluminium systems. Acta. Chim. Scand. 22: 748-752.
- LYELL, J. et al. 1964 Effects of saliva and sulfide solutions on the marginal seal of amalgam restorations. J. dent. Res. 43: 375-379.
- MANLEY, E. B. 1936 A preliminary investigation into the reaction of the pulp to various filling materials. Brit. dent. J., 60: 321-331.
- MANLEY, R. S. 1970 Adhesion in Biological Systems Academic Press - New York and London.
- MASSLER, M. & BARBER, T. K. 1953 Action of amalgam on dentin. J. Amer. dent. Ass. 47: 415-422.
- MATHEWSON, R. J. 1970 Properties of copper and silver amalgams. J. dent. Child. 37: 38-44.
- MAURICE, C. G. & SCHOUR, I. 1954 Pulpal response of the rat molar to cavity preparations and sodium fluoride. J. dent. Res. 33: 713 abstract.
- MILLER, W. D. 1890 Micro-organisms of the Human Mouth S. S. White - Philadelphia.
- MITCHELL, D. F. 1959 Irrational qualities of dental materials. J. Amer. dent. Ass. 59: 954-966.
- MORTENSEN, D. W. et al. 1965 A method of testing for marginal leakage of dental restorations with bacteria. J. dent. Res. 44: 58-63.

- MORONEY, M. J. 1956 Facts from Figures
Pelican.
- MCCUE, R. W. et al. 1951 Antibacterial properties
of some restorative
materials. Oral. Surg.
4: 1180-1184.
- MCDONALD, R. E. &
PHILLIPS, R. W. 1950 Clinical observations on a
contracting amalgam alloy.
J. dent. Res. 29: 482-485.
- McHUGH, W. D. 1955 Experiments on the hardness
and adaptation of dental
amalgam by various
condensation techniques.
Brit. dent. J. 99: 44-48.
- "NÄGELI, C. 1893 Oligodynamic phenomena
in living cells.
Denkschr. schweiz.
naturf. Ges. 33: 5-43
(Ger)
- NELSEN, R. J. et al. 1952 Fluid exchange at the
margins of dental
restorations. J. Amer.
dent. Ass. 44: 288-295.
- NOONAN, R. G. 1965 Silver amalgam is not
antibacterial. J. dent.
Child. 32: 147-153.
- PAFFENBARGER, G. C. &
DICKSON, G. 1943 Base metal alloys for
oral restorations.
J. Amer. dent. Ass.
30: 852-862.
- PAYNTER, K. J. &
WOOD, A. W. S. 1955 A method for testing
pulpal response to
stimuli using the rat
molar. J. Can. dent.
Ass. 21: 145-154.
- PHILLIPS, R. W. 1965 Cavity varnishes and
bases. Dent. Clin. N.
Amer. MAR: 159-168.
- PICKARD, H. M. &
GAYFORD, J. J. 1965 Leakage at the margins
of amalgam restorations.
Brit. dent. J. 119:
69-77.
- PREISSECKER, O. 1936 A high copper alloy
substitute for gold.
Ztschr. f. Stomatol.
34: 881 (Ger).

- RADIOLOGICAL PROTECTION
IN UNIVERSITIES P.24. 1966 The Association of
Commonwealth Univer-
sities - 36, Gordon
Square, London, W.C.1.
- RANTANEN, A. V. 1961 On the initial contrac-
tion of dental amalgams.
Odonto Revy 12: 166-173.
- RODRIGUEZ, M. S. &
DICKSON, G. 1962 Some tensile properties of
amalgam. J. dent. Res.
41: 840-852.
- ROWE, A. H. R. 1967 Reaction of the rat
molar pulp to various
materials. Brit. dent.
J. 122: 291-300.
- SCHARFENBERG, R. F. 1948 A critical appraised of the
early setting patterns
of Copper and Silver
amalgams Thesis -
Michigan.
- SCHOONOVER, I. C. &
SOUDER, W. 1941 Corrosion of dental
alloys. J. Amer. dent.
Ass. 28: 1278-1291.
- SHAY, D. E. et al. 1956 The antibacterial effects
of some dental restora-
tive materials. J. dent.
Res. 35: 25-32.
- SHEPPARD, M. S. 1935 The bacteriocidal action
of pure metals and metal
fillings. Dent. Cosmos.
77: 968-975.
- SHERMELDINE, A. B. &
SMITH, D. C. 1966 Amalgam from high and
low silver alloys.
Brit. dent. J. 121: 407-
413.
- SHROFF, F. R. 1952 Effects of filling materials
on the pulp. J. dent.
Educ. 16: 246-259.
- SKINNER, E. W. 1946 The Science of Dental
Materials 3rd ed.
Saunders - Philadelphia.
- SKINNER, E. W. &
PHILLIPS, R. W. 1967 The Science of Dental
Materials 6th ed.
Saunders - Philadelphia.

- SMITH, D. C. 1967 Amalgam from high and low silver alloys: the clinical significance of laboratory data. Brit. dent. J. 123: 94-97.
- SÖREMARK, R. et al. 1968 The penetration of metal ions from restorations into teeth. J. Pros. dent. 20: 531-540.
- STANLEY, H. R. & SWERDLOW, H. 1959 Reactions of the human dental pulp to cavity preparation. J. Prosth. dent. 9: 121-131.
- STANLEY, H. R. 1968 Design for a human pulp study (I and II). Oral Surg. 25: 633-647 and 756-764.
- STEWART, F. H. & STRADLING, G. N. 1971 Monitoring Techniques for mercury and mercury vapour in dental surgeries. Prelim. report. Brit. dent. J. 131: 299-308.
- STRELUIKINA, T. F. 1965 Copper amalgam containing tin. Stomatologiia (Moskva) 44: 27-29. (Russ)
- SUTHERS, W. D. & PARFITT, G. J. 1955 The effect in vitro of some materials used in restorative dentistry on the rate of acid production by oral bacteria. Dent. J. Aust. 27: 216-220.
- SWARTZ, M. L. & PHILLIPS, R. W. 1962 Influence of manipulative variables on the marginal adaptation of certain restorative materials. J. Prosth. dent. 12: 172-181.
- SWERDLOW, H. & STANLEY, H. R. 1962 The response of the human pulp to amalgam restorations. Oral Surg. 15: 499-508.
- TAMMAN, G. & STASSFURTH, T. H. 1925 Copper and silver amalgams. Z. Anorg. Chem. 143: 357-376.

- TAYLOR, N. O. 1929 A survey of amalgam alloys: a report to the research commission of the American dent. Ass. J. Amer. dent. Ass. 16: 590-598.
- TAYLOR, N. O. et al. 1949 The effects of variable factors on the crushing strength of dental amalgam. J. dent. Res. 28: 228-241.
- TERREY, H. & WRIGHT, C. M. 1928 The crystal structure of mercury, copper and copper amalgam. Phil. Mag. 6: 1055-1069.
- TOMES, C. 1875 On the introduction of watertight fillings - discussion. Trans. odont. Soc. 7: p.206.
- TOMES, J. 1861 Certain conditions presented by amalgams in filling faulty teeth. Brit. J. dent. Sci. 4: 295-298.
- TOVERUD, G. 1929 Mercury poisoning from amalgams. Norsk. Tandlaegeforen. Tid. (Nor)
- TRUEMAN, W. H. 1890 Copper amalgam. Dent. Cosmos. 32: p.477.
- TURKHEIM, H. J. 1953 Bacteriological investigations on dental filling materials. Brit. dent. J. 95: 1-7.
- TWEEDDALE, J. G. 1964 The Mechanical Properties of Metals. Allen & Unwin - London.
- WILSON, C. J. & RYGE, G. 1963 Clinical study of dental amalgam. J. Amer. dent. Ass. 66: 763-771.
- WILSON, C. W. 1933 Copper amalgam and high silver amalgam with reference to children's dentistry. Dent. Digest. 39: 183-185.

- WING, G. 1964 The measurement of dimensional change of dental amalgam. Aust. dent. J. 9: 426-430.
- WING, G. & LYELL, J. S. 1966 The marginal seal of amalgam restorations. Aus. dent. J. 9: 426-430.
- WING, G. & RYGE, G. 1965 Setting reactions of spherical particle amalgams. J. dent. Res. 44: 1325-1333.
- WOLCOTT, R. B. 1958 Failures in dental amalgam. J. Amer. dent. Ass. 56: 479-491.
- WORMINGTON, H. 1939 The dimensional changes of copper amalgam under various conditions of manipulation. N. West. Univ. Bull. 40: 10-15.
- WORNER, H. K. 1937 The properties of dental amalgams. Aust. J. dent. 41: 117-142.
- WORNER, H. K. 1940 The physical and mechanical properties of "copper" cements. Aust. J. dent. 44: 411-416.
- YAMAGAMI, A. & KAWAHARA, H. 1970 (abs) Cytochemical study of the toxicity of metal Hg by means of cell culture. J. dent. Res. 49: p.670
- ZANDER, H. A. 1946 The reaction of dental pulps to silicate cements. J. Amer. dent. Ass. 33: 1233-1243.